In re Application of:	BHATT	)	Examiner:	Stuart L. Hendrickson
		)		
Application No.:	10/620,269	)	Group Art Unit:	1793
		)		
Filed:	July 15, 2003	)	Confirmation No.: 8766
		)		
Docket No.:	02077 (3600-395-01)	)		

For: CARBON BLACKS AND USES THEREOF

APPEAL BRIEF
UNDER 37 C.F.R. § 41

Mail Stop **Appeal Brief — Patents**
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450   December 5, 2008

Sir:

(1)   Identification

The appellant, application, and the Examiner's identification data associated with this paper are provided in the above-captioned heading.

The appellant hereby files an Appeal Brief under 37 C.F.R. §41.37, together with the applicable fee under 37 C.F.R. §41.20(b)(2).

A Notice of Appeal under 37 C.F.R. §41.31 is being concurrently filed with the applicable fee under 41.20(b)(1).

(2)   **Table of Contents**

      (I)   ENCYCLOPEDIA OF CHEMICAL TECHNOLOGY,
           "Carbon (Carbon Black)", E.M. Dannenberg,
           Vol. 4, 4th Ed., 1992, pp. 1037-1074,
           New York, Wiley-Interscience (39 pages)

      (II)   ENCYCLOPEDIA OF INDUSTRIAL CHEMICAL ANALYSIS
           Ed. by F. Dee Snell et al., Vol. 8, pp. 163, 186-187, 243, *Interscience*
           Publishers, (6 pages).

(3)   Real Party in Interest

The real party in interest in this case is *Cabot Corporation.*

(4)   Related Appeals and Interferences

The appellant is not aware of any other appeals or interferences that will directly affect, be directly affected by, or have a bearing on the Board's decision in the present appeal.

(5)   Status of Claims

Claims 22, 23, and 25 are canceled.

Claims 1-21, 24, and 26-41 are rejected.

No claim is withdrawn.

Claims 1-21, 24, and 26-41 are on appeal.

(6)   Status of Amendments

No amendment was filed subsequent to final rejection.

(7)   Summary of Claimed Subject Matter

I.   Concise Explanation of the Subject Matter Defined in Independent Claims and Separately Argued Dependent Claims

a)   Independent Claim 1

Independent claim 1 is directed to a carbon black having an I_2 No. of from about 50 to about 112 mg/g as measured by ASTM D1510 (page 3, lines 23-24; page 17, line 18), a primary particle size of not greater than 25 nm as measured by ASTM D3849-89 (page 4, line 1; page 18, lines 4-5), and at least the following additional properties:

a)   an ash content of less than 0.1% as measured by ASTM D-1506 (page 6, lines 21-22);

b)   a total sulfur content of less than about 2% as measured by ASTM D-1619 (page 6, lines 22-23); and

c)   a 325 mesh residue of less than 20 ppm as measured by ASTM D-1514 (page 7, lines 1-2).

b)   Dependent Claim 2

Dependent claim 2, which depends from claim 1 on appeal, further specifies that the I_2 No. of the carbon black is 73-104 mg/g (page 5, line 11).

c)   Dependent Claim 11

Dependent claim 11, which depends from claim 1 on appeal, further specifies that the I_2 No. of the carbon black is 104 mg/g and the primary particle size is approximately 16 nm (page 20, lines 1-2; page 6, lines 4-9).

d) Dependent Claim 27

Dependent claim 27, which depends from claim 4/1, recites a polymer composition comprising a carbon wherein the polymer comprises LLDPE, HDPE, MDPE, or combinations thereof (page 16, lines 1-3).

e) Dependent Claim 30

Dependent claim 30, which depends from claim 29/4/1, recites an article that is a pipe, connector, cable jacketing, membrane, molding, or components thereof (page 16, lines 8-10).

f) Dependent Claim 31

Dependent claim 31, which depends from claim 29/4/1, recites an article that is a pressure pipe that comprises a polymer composition comprising the carbon black and a polymer (page 16, line 12).

g) Dependent Claim 32

Dependent claim 32, which depends from claim 29/4/1, recites a pressure pipe that is a UV pressure pipe that comprises a polymer composition comprising the carbon black and a polymer (page 3, lines 15-16; page 4, lines 19-20; page 16, lines 6-12).

h) Dependent Claim 34

Dependent claim 34, which depends from claim 1, further specifies that the carbon black has a CDBP of less than or equal to 102 cc/100 g, as measured by ASTM D3493-86 (page 4, lines 5-7, 9-11; page 5, lines 14-17).

i) Dependent Claim 37

Dependent claim 37, which depends from claim 1, further specifies that the carbon black has an I_2 No. of 50-85 mg/g; a primary particle size of less than or equal to 25 nm; and a CDBP of

less than or equal to 96 cc/100 g, as measured by ASTM D3493-6 (page 6, lines 14-16; page 17, lines 23-24).

j)   Dependent Claim 38

Dependent claim 38, which depends on claim 37/1, further specifies that the carbon black has an I_2 No. of 55-80 mg/g (page 6, lines 14-16).

k)   Dependent Claim 41

Dependent claim 41, which depends from claim 32/29/4/1, recites a carbon black forming part of a UV pressure pipe comprising at least one polymer and the carbon black has an I_2 No. of from 60-78 mg/g (page 6, lines 17-18).

(8)   Grounds of Rejection to be Reviewed on Appeal

1)   Whether claims 1-21, 24, 26-30, and 34-40 are unpatentable under 35 U.S.C. §102(b) as anticipated by U.S. Patent No. 5,877,250 to Sant (Sant '250) in view of Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60) and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).

2)   Whether claims 1-21, 24, 26-30, and 34-40 are unpatentable under 35 U.S.C. §102(b) as anticipated U.S. Patent No. 5,877,251 to Sant (Sant '251) in view of Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60) and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).

3)   Whether claims 1-21, 26-30, and 34-40 are unpatentable on the grounds of nonstatutory obviousness-type double patenting as being unpatentable over claims 22-53 of Assignee's U.S. Patent No. 6,852,790 in view of Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60) and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).

4)   Whether claims 1-10, 26, 29-33, and 41 are unpatentable on the grounds of nonstatutory obviousness-type double patenting over claims 1-21 of Assignee's U.S. Patent No. 6,482,386 in view of Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60) and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).

5)   Whether claims 1-9, 11-17, 19-21, 24-26, 28-29, and 34-36 are unpatentable under 35

U.S.C. §102(b) as anticipated by or, in the alternative, under 35 U.S.C. §103(a) as obvious over U.S.

Patent No. 6,025,429 to Yamazaki et al. in view of U.S. Patent No. 5,352,289 to Weaver et al., U.S.

Patent No. 4,755,371 to Dickerson, Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd

ed., 1946, p. 60), and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other

Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).

      6)   Whether claims 30-33 are unpatentable under 35 U.S.C. §103(a) as obvious over U.S.

Patent No. 6,025,429 to Yamazaki et al. in view of U.S. Patent No. 5,352,289 to Weaver et al., U.S.

Patent No. 4,755,371 to Dickerson, Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd

ed., 1946, p. 60), and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other

Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).

      7)   Whether claims 1-3, 11-21, 24, 34-36, and 37-40 are unpatentable under 35 U.S.C.

§102(b) as anticipated by or, in the alternative, under 35 U.S.C. §103(a) as obvious over U.S. Patent

No. 4,013,759 to Giet in view of U.S. Patent No. 4,755,371 to Dickerson, U.S. Patent No. 5,352,289

to Weaver et al., the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components

Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)), and Industrial Carbon

(Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60).

      8)   Whether claims 1-21, 24, 26-30, and 34-40 are unpatentable under 35 U.S.C. §103(a)

as obvious over U.S. Patent No. 4,013,759 to Giet in view of U.S. Patent No. 4,755,371 to

Dickerson, U.S. Patent No. 5,352,289 to Weaver et al., the Medalia article (A.I. Medalia et al.,

"Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6,

pp.481-492 (1982)), Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p.

60), and U.S. Patent No. 4,775,778 to van Konynenburg et al.

(9)   **Argument**

1.   **Rejection of Claims 1-21, 24, 26-30, and 34-40 Under 35 U.S.C. §102(b) as anticipated by Sant (U.S. Patent No. 5,877,250) in view of Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60) and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).**

Claims 1-21, 24, 26-29, 34-40

Claims 1-21, 24, 26-30, and 34-40 were finally rejected under 35 U.S.C. §102(b) as

anticipated by U.S. Patent No. 5,877,250 to Sant (Sant '250) in view of Industrial Carbon (Mantell,

C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60) and the Medalia article (A.I. Medalia et al.,

"Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6,

pp.481-492 (1982)).

The Examiner's Position

In the Final Office Action (page 2), the Examiner initially indicated that the following

characterizations of the teachings of the Industrial Carbon, the Medalia article, and the Dee Snell

article, apply to all rejections set forth in Final Office Action:

> The following applies to all rejections herein:
> The Industrial Carbon reference is presented as evidence that as-synthesized carbon blacks meet the 325 mesh limitation, except for the 'poor' grades. Note also the sulfur values reported as typical. The Medalia reference has extended discussion and pictures indicating that carbon black has a small particle size and meets the 325 mesh limitation. The concluding section teaches 1% ash as conventional. Iodine and nitrogen are shown to correlate by the Dee Snell article pg. 186 submitted.[1]

---

[1] With respect to the Dee Snell article, the appellant points out that this reference is not cited in any rejection statement in the Final Office Action. See, M.P.E.P. § 706.02(j)("Where a reference is relied on to support a rejection, whether or not in a minor capacity, that reference should be positively included in the statement of the rejection. See *In re Hoch*, 428 F.2d 1341, 1342 n.3 166 USPQ 406, 407 n. 3 (CCPA 1970)").

According to the Final Office Action (page 2), the Examiner interprets and applies Sant '250 in view of Industrial Carbon and the Medalia article to claims 1-21, 24, 26-30, and 34-40 in the following manner:

> The [Sant '250] reference teaches what appears to be the claimed carbon black. Specification pg. 13 indicates that the present carbon black is the same as that of Sant, with no modifications made.

The Examiner added the following remarks in the Final Office Action (pages 4-5) in support of this rejection:

> ... The arguments concerning the Sant references rely upon semantics in the specification. No patentable difference is seen between 'carbon black' and 'carbon black products' since carbon blacks are decomposition products. Given that the references belong to the applicant, it is not seen why an analysis could not be undertaken. ... Note that that ['386] patent also refers to Sant '250 as the basis for its synthesis, just like the present specification does. This is evidence that the carbon blacks do not patentably differ. ...

For the following reasons, the appellant requests review and reversal of this rejection.

The Appellant's Position

The present invention is directed to carbon blacks, and polymer compositions and articles that incorporate these carbon blacks, wherein the carbon blacks have an I_2 No. of from about 50 to about 112 mg/g, primary particle size of not greater than 25 nm, and at least the following properties: a) an ash content of less than 0.1%; b) a total sulfur content of less than about 2%; and c) a 325 mesh residue of about 20 ppm or less (page 3, lines 14-16; page 3, line 23 to page 4, line 4; page 4, lines 16-17; page 6, line 20 to page 7, line 3). The carbon blacks that combine these characteristics have appropriate properties for use in polymeric compositions such as UV applications like pipe, film, membranes, jacketing, and the like (page 3, lines 14-16; page 4, lines 18-20). An advantage of the carbon blacks of the present invention is that the carbon blacks preferably

impart low viscosity to the polymer compositions into which they are incorporated (page 16, lines 16-18). Another advantage of the carbon blacks of the present invention is that the carbon blacks impart low CMA (compound moisture absorption) to the polymer compositions into which they are incorporated (page 16, lines 19-21). A further advantage of the carbon blacks of the present invention is that the carbon blacks can be incorporated at high carbon black loadings into polymer compositions (page 16, lines 22-24).

The reference to Sant '250 at page 13 of the present application is with respect to the processes that can be used to make "carbon black products" in general. The reference to "carbon black products" at page 13 is simply with respect to the raw materials, as well as the products that can incorporate carbon black, such as the polymer compositions. It is respectfully noted that the term "carbon black <u>products</u>" is referred to when the present application refers at page 13 to Sant '250, and not the term "carbon blacks." In view of this clarification, Sant '250 does not teach or suggest the carbon blacks of the present invention as set forth, for instance, in claim 1 and the dependent claims. No admission was made that the Sant '250 patent teaches or suggests the claimed invention.

Sant '250 also does not teach or suggest an ash content, sulfur content, or a 325 mesh residue as recited in the claims of the present application (e.g., see Examples 1-14, Table 3, at cols. 14-15). The Medalia article cited by the Examiner clearly and unequivocally teaches that a number of prior carbon blacks all had an ash content *significantly exceeding* 0.1%. Reference is made in this respect to carbon black samples #375, RC (Fluffy), N351, N762, N472 and NC in Table 1 on page 489 of the Medalia article. As indicated in Table 1 of the Medalia article, these prior carbon blacks had original ash content ranging from 0.27 to 0.92%. These ash contents are approximately 300% higher or more than the ash content of less than 0.1% specified for the presently claimed carbon black. Therefore,

these conventional carbon black values evidenced by Medalia do not anticipate or suggest the much lower ash content and sulfur content for the carbon blacks of the present invention, which are also provided at the prescribed primary particle size range and iodine number value range.

The Industrial Carbon reference fails to make up for this difference, as it has nothing to say about ash content or iodine values of carbon blacks. Industrial Carbon refers to sulfur content that ordinarily can be low as 0.01 to 0.03 but that occasionally values as high as 0.4% are encountered. The Industrial Carbon reference also specifically states that carbon blacks leave traces on the 325 mesh screen and the weight percents can be from 0.03 to 0.15 percent. 0.03 percent equates to 300 ppm, which is outside of the range recited in claim 1, which is a 325 mesh residue of less than 20 ppm. Further, the reference in Industrial Carbon to carbon black "traces" left on 325 mesh screen (44 micron) on screen analysis, or, in poorer grades, 0.03 to 0.15%, is understood to be an independent parameter from ash content. In addition, Industrial Carbon refers to carbon blacks having an estimated diameter of from 60 to less than 20 millimicrons. However, the Industrial Carbon reference nowhere suggests that ash content and iodine values can be predicted from sulfur content, 325 mesh and/or particle sizes that might known for a given carbon black. Further, there is no teaching, motivation, or prediction of success for one skilled in the art to make the carbon blacks having the particular combined parameters of claim 1.

In view of the above facts and evidence, this anticipation rejection is inappropriate since the Examiner is relying on multiple references, inclusive of Industrial Carbon and the Medalia article, to reject these claims under an anticipation rejection in a manner that does not comport with "modest flexibility" allowed under certain circumstances to the all elements rule of anticipation as discussed in M.P.E.P. §2131.01, III. In relevant part, M.P.E.P. §2131.01, III, instructs as follows:

> "To serve as an anticipation when the reference is silent about the asserted inherent characteristic, such gap in the reference may be filled with recourse to extrinsic evidence. Such evidence must make clear that the missing descriptive matter is necessarily present in the thing described in the reference, and that it would be so recognized by persons of ordinary skill." *Continental Can Co. USA v. Monsanto Co.,* 948 F.2d 1264, 1268, 20 USPQ2d 1746, 1749 (Fed. Cir. 1991)(The court went on to explain that "this modest flexibility in the rule that 'anticipation' requires that every element of the claims appear in a single reference accommodates situations in which the common knowledge of technologists is not recorded in the reference; that is, where technological facts are known to those in the field of the invention, albeit not known to judges." 948 F.2d at 1268, 20 USPQ at 1749-50.). ...

However, in view of the reasons explained above, the Industrial Carbon reference and the Medalia article are not evidence that makes clear that the ash content and 325 mesh residue properties that are presently recited are *necessarily present* in the carbon black disclosed by Sant '250, and that it would be so recognized by persons of ordinary skill.

In view of the above-identified differences between the present claim 1 on appeal and Sant '250, Industrial Carbon, and Medalia, it is apparent that the Examiner has not established a prima facie case of anticipation (or obviousness) against present claim 1. Therefore, no experimental analysis should be required of appellant to show the presently carbon blacks are not inherent or obvious over a carbon black disclosed by Sant '250.

In view of the above differences, and contrary to the assertion made in the Final Office Action (page 2), the Examiner has not found in the art "what appears to be the claimed carbon black" as presently claimed. In relevant part, M.P.E.P. § 2112, IV, instructs as follows:

> " ... Inherency, however, may not be established by probabilities or possibilities. The mere fact that a certain thing may result from a given set of circumstances is not sufficient." *In re Robertson*, 169 F.3d 743, 745, 49 USPQ2d 1949, 1950-51 (Fed. Cir. 1999) (citations omitted) ...

In view of the above explanations by the appellant, it is apparent that the Examiner has attempted to premise an anticipation rejection of claim 1 on appeal based on mere probabilities or possibilities, which is improper.

Claims 2-21, 24, 26-30, and 34-40, which ultimately depend from claim 1, are not anticipated by nor obvious over Sant '250, Industrial Carbon, or the Medalia article, for at least the same reasons as set forth above relative to their parent claim 1, and reference is made thereto.

In view of the above, the appellant requests reversal of the rejection of claims 1-21, 24, 26-30, and 34-40 on appeal based on Sant '250, Industrial Carbon, or the Medalia article.

For these reasons, this rejection should be reversed.

Claim 30

Dependent claim 30, which depends from claim 29/4/1, recites an article that is a pipe, connector, cable jacketing, membrane, molding, or components thereof, wherein the article comprises a polymer composition comprising at least one polymer and the carbon black of claim 1.

The reasons for reversal as discussed above with respect to the rejection of parent claim 1 based on Sant '250, Industrial Carbon, and Medalia apply equally to claim 30. Additional reasons for reversal are as follows.

Sant '250 refers to polymer compositions in general. Sant '250 does not teach or suggest any of the articles recited in present claim 30.

In view of the above reasons, this rejection of claim 30 should be reversed.

2. **Rejection of Claims 1-21, 24, 26-30, and 34-40 under 35 U.S.C. §102(b) as anticipated by Sant (U.S. Patent No. 5,877,251) in view of Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60) and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).**

Claim 1-21, 24, 26-29, and 34-40

Claims 1-21, 24, 26-30, and 34-40 were finally rejected under 35 U.S.C. §102(b) as anticipated by U.S. Patent No. 5,877,251 to Sant (Sant '251) in view of Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60) and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).

The Examiner's Position

According to the Final Office Action (page 2), the Examiner interprets and applies Sant '251 in view of Industrial Carbon and the Medalia article to claims 1-21, 24, 26-30, and 34-40 in the following manner:

> The [Sant '251] reference teaches what appears to be the claimed carbon black. Specification pg. 13 indicates that the present carbon black is the same as that of Sant, with no modifications made.

The Examiner added the following remarks, in relevant part, in the Final Office Action (pages 4-5) in support of this rejection:

> ... The arguments concerning the Sant references rely upon semantics in the specification. No patentable difference is seen between 'carbon black' and 'carbon black products' since carbon blacks are decomposition products. Given that the references belong to the applicant, it is not seen why an analysis could not be undertaken. ...

For the following reasons, the appellant requests review and reversal of this rejection.

The Appellant's Position

For the same reasons as explained above by the appellant with respect to Sant '250, the reference to Sant '251 at page 13 of the present application is with respect to the processes that can be used to make "carbon black products" in general, and not carbon blacks such as presently claimed.

No admission was made that the Sant '251 patent teaches or suggests the claimed invention.

Sant '251 also does not teach or suggest an ash content, sulfur content, or a 325 mesh residue as recited in the claims of the present application (e.g., see Examples 1-33, Table 3, at cols. 28-30). As indicated, the Medalia article cited by the Examiner clearly and unequivocally teaches that a number of prior carbon blacks all had ash content *significantly exceeding* 0.1%. As also indicated, the Industrial Carbon reference fails to make up for this difference, as it has nothing to say about ash content or iodine values of carbon blacks. Also, the Industrial Carbon reference nowhere suggests that ash content and iodine values can be predicted from sulfur content, 325 mesh and/or particle sizes that might known for a given carbon black. Further, there is no teaching, motivation, or prediction of success for one skilled in the art to make the carbon blacks having the particular combined parameters of claim 1.

Claims 2-21, 24, 26-30, and 34-40, which ultimately depend from claim 1, are not anticipated by nor obvious over Sant '251, Industrial Carbon, or the Medalia article, for at least the same reasons as set forth above relative to their parent claim 1, and reference is made thereto.

In view of the above, the appellant requests reversal of the rejection of claims 1-21, 24, 26-30, and 34-40 on appeal based on Sant '251, Industrial Carbon, or the Medalia article.

For these reasons, this rejection should be reversed.

Claim 30

Dependent claim 30, which depends from claim 29/4/1, recites an article that is a pipe, connector, cable jacketing, membrane, molding, or components thereof, wherein the article comprises a polymer composition comprising at least one polymer and the carbon black of claim 1.

The reasons for reversal as discussed above with respect to the rejection of parent claim 1

- 19 -

based on Sant '251, Industrial Carbon, and Medalia apply equally to claim 30. Additional reasons

for reversal are as follows.

Sant '251 refers to polymer compositions in general. Sant '251 does not teach or suggest

any of the articles recited in present claim 30.

In view of the above reasons, this rejection of claim 30 should be reversed.

3. **Rejection of Claims 1-21, 26-30, and 34-40 Under the Judicially Created Doctrine of Obviousness-type Double Patenting Over Claims 22-53 of Assignee's U.S. Pat. No. 6,852,790 in view of Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60) and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).**

<u>Claims 1-21, 26-30, 34-36, 39, and 40</u>

Claims 1-21, 26-30, and 34-40 were finally rejected on the grounds of nonstatutory

obviousness-type double patenting as being unpatentable over claims 22-53 of Assignee's U.S.

Patent No. 6,852,790 in view of Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed.,

1946, p. 60) and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components

Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).

The Examiner's Position

According to the Final Office Action (page 2), the Examiner interprets and applies

assignee's U.S. Pat. No. 6,852,790 patent in view of Industrial Carbon and the Medalia article to

claims 1-21, 26-30, and 34-40 in the following manner:

> Although the conflicting claims are not identical, they are not patentably distinct from each other because they claim common, overlapping, subject matter in the ranges of values.

> The '790 patent elucidates in col. 3 overlapping particle size, and other properties. The polymers are indicated in col. 8.

The Examiner also added the following remarks in the Final Office Action (page 5) in support of the rejection:

> Concerning the ODP over '790, it is argued that the specification thereof cannot be relied upon. However, this is not true, because inherent properties are being relied upon. ... Since '790 and '386 are both owned by the applicant, it is not seen why an analysis could not be offered to demonstrate the properties of these carbons. ...

For the following reasons, the appellant requests review and reversal of this rejection.

The Appellant's Position

Appellant submits that the Examiner's reasoning is flawed in his assertion that the present claims 1-21, 26-30, and 34-40 are obvious over claims 22-53 of Assignee's U.S. Patent No. 6,852,790. The analysis of possible obviousness double patenting is focused on whether "conflicting claims" are present between the instant application and the previously issued patent to assignee, and not whether there may be a conflicting claim when combined with unclaimed specification disclosures, as suggested in the Office Action (e.g., see M.P.E.P. § 804 *et seq.*). As explained in M.P.E.P. 804, B.,1.:

> When considering whether the invention defined in a claim of an application would have been an obvious variation of the invention defined in the claim of a patent, the disclosure of the patent may not be used as prior art. *General Foods Corp. v. Studiengesellschaft Kohle mbH*, 972 F.2d 1272, 1279, 23 USPQ2d 1839, 1846 (Fed. Cir. 1992). This does not mean that one is precluded from all use of the patent disclosure.
>
> The specification can be used as a dictionary to learn the meaning of a term in the patent claim. [citation omitted] ... ... Further, those portions of the specification which provide support for the patent claims may also be examined and considered when addressing the issue of whether a claim in the application defines an obvious variation of an invention claimed in the patent. [citation omitted] ...

Therefore, in asserting an obviousness-type double patenting rejection, the specification can only be referenced under very limited circumstances, viz., use of the specification as a dictionary or the claim supporting portions of the patent specification indicate whether the application claim on appeal is an obvious variation of an invention claimed in the patent. The Examiner ostensibly does not refer to the supporting portions of the specification of U.S. Pat. No. 6,852,790 as a dictionary, so the reference ostensibly must be premised by the Examiner on an assertion that the supporting portions of its specification teach that unclaimed features are "obvious variations" to the different ones that are claimed in the given patent.

In this respect, the alleged elucidation in column 3 of U.S. Patent No. 6,852,790 of overlapping particle sizes focuses solely on one possible overlapping parameter but does not address whether Assignee's U.S. Pat. No. 6,852,790 claims a carbon black having overlapping values with respect to *all* properties recited for the carbon black of claim 1 on appeal. As can be appreciated, the claimed carbon black is defined by its entire set of recited properties in combination, and not the properties individually in isolation from one another. U.S. Pat. No. 6,852,790 does not teach a carbon black, as a whole, that overlaps with that claimed on appeal. The appellant points out that U.S. Pat. No. 6,852,790 fails to claim or teach in the supporting portions of the specification any values for sulfur contents, 325 mesh residues, or ash contents of the carbon black product. Sulfur amounts of preferably less than about 8000 ppm are disclosed in U.S. Pat. No. 6,852,790 for the *feedstock*, but not for the carbon product *per se* (col. 6, lines 25-26). Therefore, when comparing the claims of the present application with the claims of U.S. Pat. No. 6,852,790, the presently claimed carbon black, with *all* its recited features being considered in combination, is not claimed by U.S. Pat. No. 6,852,790, nor is it disclosed to be an obvious variation in the supporting portions of the

specification of the patent to the compositions as claimed in U.S. Pat. No. 6,852,790.

Therefore, this obviousness-type double patent rejection is improper.

The Examiner also references the Industrial Carbon and Medalia references. However, as explained above, these references differ from the present claims, and reference is made to those previous explanations which also apply here, and the Industrial Carbon and the Medalia references would not make up for the differences between the present claims and the claims of U.S. Pat. No. 6,852,790 patent. For example, and as explained above with respect to the Industrial Carbon reference, this reference specifically states that carbon blacks leave traces on the 325 mesh screen and the weight percents can be from 0.03 to 0.15 percent. 0.03 percent equates to 300 ppm, which is outside of the range recited in claim 1 on appeal, which is a 325 mesh residue of less than 20 ppm. Again, neither the claims nor specification of assignee's U.S. Pat. No. 6,852,790 make reference to 325 mesh residues, sulfur levels, or ash contents.

In view of at least the above differences, none of claims 1-21, 26-30, and 34-40 are obvious over claims 22-53 of Assignee's U.S. Patent No. 6,852,790.

For at least these reasons, this obviousness-type double patent rejection of claims 1-21, 26-30, and 34-40 on appeal based on U.S. Pat. No. 6,852,790 is improper and should be reversed.

Claim 37

Claim 37 further recites carbon black having an I_2 No. of 50-85 mg/g; a primary particle size of less than or equal to 25 nm; and a CDBP of less than or equal to 96 cc/100 g.

The reasons for reversal as discussed above with respect to the rejection of parent claim 1 based on U.S. Pat. No. 6,852,790, Industrial Carbon, and Medalia apply equally to claim 37. Additional reasons for reversal are as follows.

No <u>explicit</u> analysis is provided in the final rejection explaining which claim or claims of U.S. Patent No. 6,852,790 recite(s) a carbon black meeting all recitations of claim 37 on appeal, and none is found based on the appellant's review. *KSR Int'l Co. v. Teleflex Inc.*, 127 S.Ct. 1727, 1740-41, 82 USPQ2d 1385, 1396 (2007)(internal cite omitted). For instance, there is an absence in the Final Office Action of an "... identified reason that would have prompted a person of ordinary skill in the relevant field to combine the elements in the way the claimed invention does" *Id.*, 127 S.Ct. at 1741, 82 USPQ2d at 1396. Therefore, there is insufficient analysis set forth by the Examiner in the Final Office Action to support a *prima facie* case of obviousness against dependent claim 37 on appeal based on the claims of U.S. Pat. No. 6,852,790.

This obviousness-type double patent rejection of claim 37 on appeal based on U.S. Pat. No. 6,852,790 is improper and should be reversed.

Claim 38

Claim 38, which depends on claim 37, further recites carbon black wherein the I_2 No. is 55-80 mg/g. The reasons for reversal as discussed above with respect to the rejection of parent claim 1 based on U.S. Pat. No. 6,852,790, Industrial Carbon, and Medalia apply equally to claim 38. Additional reasons for reversal are as follows.

No explicit analysis is provided in the rejection explaining which claim or claims of U.S. Patent No. 6,852,790 recite(s) a carbon black meeting all recitations of claim 38 on appeal, and none is found based on the appellant's review.

This obviousness-type double patent rejection of claim 38 on appeal based on U.S. Pat. No. 6,852,790 is improper and should be reversed.

4. **Rejection of Claims 1-10, 26, 29-33, and 41 Under the Judicially Created Doctrine of Obviousness-type Double Patenting Over Claims 1-21 of Assignee's U.S. Pat. No. 6,482,386 in view of Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60), and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black,"** *Carbon,* **Vol. 20, No. 6, pp.481-492 (1982)).**

<u>Claims 1-10, 26, 29-30, 33</u>

Claims 1-10, 26, 29-33, and 41 were finally rejected on the grounds of nonstatutory

obviousness-type double patenting over claims 1-21 of Assignee's U.S. Patent No. 6,482,386 in view

of Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60) and the Medalia

article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black,"

Carbon, Vol. 20, No. 6, pp.481-492 (1982)).

The Examiner's Position

According to the Final Office Action (page 3), the Examiner interprets and applies

assignee's U.S. Pat. No. 6,482,386 patent in view of Industrial Carbon and the Medalia article to

claims 1-10, 26, 29-33, and 41 in the following manner:

> Although the conflicting claims are not identical, they are not patentably
> distinct from each other because the numerical values overlap. Column 4
> teaches a tube shape and fluffy form, which indicates the 325 mesh
> limitation is met.

The Examiner also added the following remarks in the Final Office Action (page 5) in

support of the rejection:

> Concerning the ODP over '790, it is argued that the specification thereof
> cannot be relied upon. However, this is not true, because inherent
> properties are being relied upon. The argument concerning '386 is similar.
> Note that that patent also refers to Sant '250 as the basis for its synthesis,
> just like the present specification does. This is evidence that the carbon
> blacks do not patentably differ. Since '790 and '386 are both owned by
> the applicant, it is not seen why an analysis could not be offered to

demonstrate the properties of these carbons.

For the following reasons, the appellant requests review and reversal of this rejection.

The Appellant's Position

As indicated above, the analysis of possible obviousness double patenting is focused on whether "conflicting claims" are present between the instant application and the previously issued patent to assignee, and not whether there may be a conflicting claim when combined with unclaimed specification disclosures, as suggested in the Final Office Action (e.g., see M.P.E.P. § 804 *et seq.*). The appellant submits that the Examiner's proposition is improper as applied to the present circumstances for the reasons explained above in connection with discussion of the rejection based on U.S. Pat. No. 6,852,790, and reference is made thereto. Namely, U.S. Pat. No. 6,482,386 also fails to claim, or teach in the supporting portions of the specification, any values for sulfur contents, 325 mesh residues, or ash contents. Sulfur amounts of preferably less than about 1000 ppm are disclosed in U.S. Pat. No. 6,482,386 for the *feedstock*, but not for the carbon product *per se* (col. 4, lines 4-5). Therefore, when comparing the claims of the present application with the claims of U.S. Pat. No. 6,482,386, the presently claimed carbon black, with *all* its recited features being considered in combination, is not claimed by U.S. Pat. No. 6,482,386, nor is it disclosed to be an obvious variation in the supporting portions of the specification of the patent to the compositions as claimed in U.S. Pat. No. 6,482,386.

With respect to the assertion in the Final Office Action that the teaching of a tube shape or fluffy form at column 4 of U.S. Pat. No. 6,482,386 indicates the 325 mesh limitation for the carbon black of claim 1 on appeal is met, the appellant disagrees because this assertion by the Examiner is based on a misreading of the patent reference or fails to explain a nexus between these shapes and

the 325 mesh recitation of the carbon black recited in claim 1 on appeal. In fact, column 4, lines 51-65 of U.S. Pat. No. 6,482,386 does *not* refer to tube-shaped carbon black shapes, but instead refers to tube shapes of *extruded articles* made with polymeric compositions containing the carbon black. Column 4, lines 16-19 of U.S. Pat. No. 6,482,386 refer to fluffy or pelletized forms of carbon black, but the Examiner does not provide an explicit analysis of any connection alleged to inherently arise between either of such morphologies and the 325 mesh limitation recited in claim 1 on appeal.

The Examiner also references the Industrial Carbon and Medalia references. However, these references differ from the present claims as explained above, and reference is made thereto, and they would not make up for the differences between the present claims and the claims of the '386 patent. For example, and as explained above with respect to the Industrial Carbon reference, this reference specifically states that carbon blacks leave traces on the 325 mesh screen and the weight percents can be from 0.03 to 0.15 percent. 0.03 percent equates to 300 ppm, which is outside of the range recited in claim 1 on appeal, which is a 325 mesh residue of less than 20 ppm. Again, neither the claims nor specification of assignee's U.S. Pat. No. 6,482,386 make reference to 325 mesh residues, sulfur levels, or ash contents.

With respect to the Examiner's comment at page 5 of the Final Office Action that assignee's U.S. Pat. No. 6,482,386 also makes reference to the Sant '250 reference, the appellant again explains that the term "carbon black <u>products</u>" is referred to when the present application refers at page 13 to Sant '250, and not the term "carbon blacks." U.S. Pat. No. 6,482,386 appears to refer to "carbon blacks" of Sant '250 at column 3, line 43 to column 4, line 12. In view of this clarification, U.S. Pat. No. 6,482,386, and inclusive of any internal reference therein to Sant '250, does not teach

or suggest the carbon blacks of the present invention as set forth, for instance, in claim 1 and the

dependent claims. Again, no admission was made that the Sant '250 patent or '251 patent teaches or

suggests the claimed invention.

In view of at least the above differences, none of claims 1-10, 26, 29-33 and 41 are obvious

over claims 1-21 of Assignee's U.S. Patent No. 6,482,386.

For at least these reasons, this obviousness-type double patent rejection of claims 1-10, 26,

29-33 and 41 on appeal based on U.S. Pat. No. 6,482,386 is improper and should be reversed.

Claim 31

Claim 31 recites an article that is a pressure pipe that comprises a polymer composition

comprising the carbon black and a polymer. The reasons for reversal as discussed above with

respect to the rejection of parent claim 1 based on U.S. Pat. No. 6,482,386, Industrial Carbon, and

Medalia apply equally to claim 31. Additional reasons for reversal are as follows.

No explicit analysis is provided in the rejection explaining which claim or claims of U.S.

Patent No. 6,482,386 recite(s) a pressure pipe meeting all recitations of claim 31 on appeal, and none

is found based on the appellant's review.

This obviousness-type double patent rejection of claim 31 on appeal based on U.S. Pat. No.

6,482,386 is improper and should be reversed.

Claim 32

Claim 32 recites a pressure pipe that is a UV pressure pipe that comprises a polymer

composition comprising the carbon black and a polymer. The reasons for reversal as discussed

above with respect to the rejection of parent claim 1 based on U.S. Pat. No. 6,482,386, Industrial

Carbon, and Medalia apply equally to claim 32. Additional reasons for reversal are as follows.

No explicit analysis is provided in the rejection explaining which claim or claims of U.S. Patent No. 6,482,386 recite(s) a UV pressure pipe meeting all recitations of claim 32 on appeal, and none is found based on the appellant's review.

This obviousness-type double patent rejection of claim 32 on appeal based on U.S. Pat. No. 6,482,386 is improper and should be reversed.

Claim 41

Claim 41 recites a carbon black forming part of a UV pressure pipe comprising at least one polymer and the carbon black which has an I_2 No. of from 60-78 mg/g, primary particle size of not greater than 25 nm, and at least the following properties: a) an ash content of less than about 1%; b) a total sulfur content of less than about 2%; and c) a 325 mesh residue of about 200 ppm or less. The reasons for reversal as discussed above with respect to the rejection of parent claim 1 based on U.S. Pat. No. 6,482,386, Industrial Carbon, and Medalia apply equally to claim 41. Additional reasons for reversal are as follows.

No explicit analysis is provided in the rejection explaining which claim or claims of U.S. Patent No. 6,482,386 recite(s) a UV pressure pipe meeting all recitations of claim 41 on appeal, and none is found based on the appellant's review.

This obviousness-type double patent rejection of claim 41 on appeal based on U.S. Pat. No. 6,482,386 is improper and should be reversed.

5. **Rejection of Claims 1-9, 11-17, 19-21, 24-26, 28, 29, and 34-36 Under 35 U.S.C. § 102(b), or in the alternative, under 35 U.S.C. § 103(a) Over Yamazaki et al. (U.S. Pat. No. 6,025,429) in view of Weaver et al. (U.S. Pat. No. 5,352,289), Dickerson (U.S. Pat. No. 4,755,371), Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60), and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And**

Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).

Claims 1-9, 12-17, 19-21, 24-26, 28, 29, and 34-36

Claims 1-9, 11-17, 19-21, 24-26, 28-29, and 34-36 were finally rejected under 35 U.S.C.

§102(b) as anticipated by or, in the alternative, under 35 U.S.C. §103(a) as obvious over U.S. Patent

No. 6,025,429 to Yamazaki et al. in view of U.S. Patent No. 5,352,289 to Weaver et al., U.S. Patent

No. 4,755,371 to Dickerson, Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed.,

1946, p. 60), and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components

Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).

The Examiner's Position

According to the Final Office Action (page 3), the Examiner interprets and applies

Yamazaki et al., Weaver et al., Dickerson, Industrial Carbon, and the Medalia article to claims 1-9,

11-17, 19-21, 24-26, 28, 29, and 34-36 in the following manner:

> Yamazaki teaches in the entire document, especially ex. 6 and col. 11,
> examples of acetylene blacks having low ash and grit contents, and iodine
> values of 92 and 110. Col. 1 teaches polymers and col. 3 teaches the
> loading. This does not explicitly teach all the claimed properties, however
> Weaver teaches in col. 3 that acetylene blacks are known to be low in ash
> and S, which is expected because they are made from a source which
> contains little or no S or metals. Note also the DBP values recited.
> Dickerson teaches in column 6 the claimed 325 mesh residue for carbon
> black. The Industrial Carbon reference is presented as evidence that as-
> synthesized carbon blacks meet the 325 mesh limitation. Therefore, it
> appears based upon this additional evidence that the carbon black of
> Yamazaki renders the claims unpatentable. The particle size is unknown,
> but a difference should be shown.

The Examiner also added the following remarks in the Final Office Action (page 5) in

support of the rejection:

> ... Concerning Yamazaki, the ash (and sulfur) limitation does not

distinguish, since the reference does not uses oils (which are known to contain ash and S compounds) but rather gases. What Medalia teaches about normal ash contents of oil blacks is irrelevant. To amplify this point, it is noted that gas supplied to ordinary homes is so pure that the gas company deliberately adds mercaptan (S compound) in trace amounts to permit it to be detected by smell.

For at least the following reasons, the appellant requests review and reversal of this rejection.

The Appellant's Position

As previously pointed, the present claims recite, *inter alia*, an ash content of less than 0.1%, not 1%. In addition, with respect to the Examiner's reliance on Yamazaki et al., it is respectfully noted that the grit content and ash content of Yamazaki et al. are not based on ASTM standards, but instead are based on JIS standards, as indicated at col. 7, lines 36-55. It is respectfully noted that the JIS standards are not the same as the ASTM standards relied upon in the present application. There is no evidence of record that measurement values obtained for grit content and ash content using the JIS standards are predictive in a known manner of results that would be obtained by ASTM standards. As indicated above, the various properties of the present invention are determined using ASTM test procedures and this was specifically indicated at pages 5-7 of the present application and elsewhere. Accordingly, reliance on these numbers in the Examiner's final rejection without addressing how these numbers would correspond to ASTM standards fails to establish a *prima facie* case of obviousness or even anticipation.

In addition, it is respectfully noted that Weaver et al., while discussing carbon blacks, primarily relates to furnace carbon blacks, but does compare furnace carbon blacks to acetylene carbon blacks. It is further noted that the ash content and sulfur content mentioned in Weaver et al.

are not based on an ASTM standard. The Examiner has not explained how the particular standards used in Weaver et al. would compare to the ASTM standards used in the present application.

With respect to the Examiner's reliance on Dickerson, the Examiner's reliance on the claimed 325 mesh is with respect to furnace carbon blacks and, therefore, its combination with Yamazaki et al. would be difficult at best. Merging acetylene carbon black properties with furnace carbon black properties simply is not a combination that one skilled in the art would make since the carbon blacks are made differently and have different properties. Even one reference relied upon by the Examiner, Weaver et al., acknowledges the difference in acetylene carbon blacks and furnace carbon blacks (e.g., col. 3, lines 23-47). Also, it appears that Weaver et al. does not use an ASTM standard to measure the 325 mesh and, therefore, has the same problems as the above-mentioned references.

In addition, Weaver et al. fails to teach any I_2 No. or primary particle sizes of acetylene blacks. Dickerson refers to "Feedstock Oil" in the referenced EXAMPLE, and clearly does not relate to acetylene blacks (col. 5, line 49, col. 6, line 50), and thus Dickerson fails to possibly teach any "inherent" properties of the acetylene black of Yamazaki et al.

The Examiner also refers to the Industrial Carbon and Medalia references. However, these references differ from the present claims, as explained above, and they would not make up for the differences between the present claims and Yamazaki et al., Weaver et al., and Dickerson. For example, as explained above with respect to the Industrial Carbon reference, this reference relied upon by the Examiner does not provide a 325 mesh limitation of 200 ppm or less. In fact, the only number provided is a weight percent level of from 0.03 to 0.15%, which would be far above a 20 ppm level. As indicated above, 0.03 percent equates to 300 ppm, which is outside the range recited in claim 1 on appeal. Further, Industrial Carbon have nothing to say about acetylene blacks in

particular, and, therefore, it fails to possibly teach any "inherent" properties of the acetylene black of Yamazaki et al.

In the Final Office Action (page 5), the Examiner suggested that what Medalia teaches about normal ash contents of oil blacks is irrelevant, noting that Yamazaki et al. uses gases and not oils as feedstock, and that gas supplied to ordinary homes is so pure that the gas company adds scent to it. In response, the appellant points out that the Examiner can not arbitrarily pick and choose from the Medalia reference that which is urged by the Examiner to support the Examiner's rejection, such as small particle size and the 325 mesh limitation, while ignoring other essential features of the carbon blacks described in the Medalia article that are necessary to the full appreciation of what the Medalia reference fairly suggests to one of ordinary skill in the art. *In re Wesslau*, 353 F.2d 238, 241, 147 USPQ 391, 393 (CCPA 1965). In addition, Yamazaki et al. teaches thermal decomposition of "acetylene", as the feedstock material, to form acetylene black feed powder feed, which in turn is granulated (see col. 3, lines 46-49; col. 9, lines 10-35). Acetylene is <u>not</u> methane gas. Methane gas is the gas ordinarily supplied to homes. There is no factual evidence of record showing that the purity of methane gas supplied to homes is predictive of the purity of acetylene used in industrial applications, such as that disclosed by Yamazaki et al. Furthermore, there is no factual evidence of record that shows that the profile of decomposition products of methane gas is predictive of those of acetylene gas such as used in Yamazaki et al.

In addition, claim 1 and the claims dependent on claim 1 <u>all require</u> a carbon black having a <u>primary particle size of not greater than 25 nm</u>. Yamazaki et al., Weaver et al., Dickerson, the Industrial Carbon, and the Medalia article do not teach or even mention a primary particle size. This point was not addressed in this rejection as set forth in the Final Office Action.

Claims 2-9, 11-17, 19-21, 24-26, 28, 29, and 34-36, which ultimately depend from claim 1, are not anticipated by nor obvious over Yamazaki et al., Weaver et al., Dickerson, Industrial Carbon, and the Medalia article for at least the same reasons as set forth above relative to their parent claim 1.

In view of at least the above reasons, this rejection of claims 1, 2-9, 11-17, 19-21, 24-26, 28, 29, and 34-36, on appeal based on Yamazaki et al., Weaver et al., Dickerson, the Industrial Carbon, and the Medalia article should be reversed.

Claim 11

Claim 11 further specifies an even smaller primary particle size of approximately 16 nm, in combination with an I_2 No. value of approximately is 104 mg/g. The reasons for reversal as discussed above with respect to the rejection of parent claim 1 based on Yamazaki et al., Weaver et al., Dickerson, the Industrial Carbon reference, and the Medalia article, apply equally to claim 11. Additional reasons for reversal are as follows.

The recitation in claim 11 of an even smaller primary particle size of approximately 16 nm, in combination with an I_2 No. value of approximately is 104 mg/g is even further distanced from Yamazaki et al., Weaver et al., Dickerson, the Industrial Carbon reference, and the Medalia article, which do not teach or even mention a primary particle size.

Carbon blacks of claim 11 on appeal are not anticipated by or obvious over Yamazaki et al., Weaver et al., Dickerson, the Industrial Carbon reference, and the Medalia article.

In view of the above, reversal of the rejection of claim 11 on appeal based on Yamazaki et al., Weaver et al., Dickerson, the Industrial Carbon reference and the Medalia article is respectfully requested.

6. **Rejection of Claims 30-33 Under § 103(a) Over Yamazaki et al. (U.S. Pat. No. 6,025,429) in view of Weaver et al. (U.S. Pat. No. 5,352,289), Dickerson (U.S. Pat. No. 4,755,371), Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60), and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).**

<u>Claim 30</u>

Claims 30-33 were finally rejected under 35 U.S.C. §103(a) as obvious over U.S. Patent No. 6,025,429 to Yamazaki et al. in view of U.S. Patent No. 5,352,289 to Weaver et al., U.S. Patent No. 4,755,371 to Dickerson, Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60), and the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)).

The Examiner's Position

According to the Final Office Action (pages 3-4), the Examiner interprets and applies Yamazaki et al., Weaver et al., Dickerson, Industrial Carbon, Industrial Carbon, and the Medalia article to claims 30-33 in the following manner:

> The references do not teach the claimed pipe, however forming one is an obvious expedient to create an useful artifact made of the carbon and polymer with the desired properties.

The Examiner also added the following remarks in the Final Office Action (page 5) in support of the rejection:

> ... Concerning Yamazaki, the ash (and sulfur) limitation does not distinguish, since the reference does not uses oils (which are known to contain ash and S compounds) but rather gases. What Medalia teaches about normal ash contents of oil blacks is irrelevant. To amplify this point, it is noted that gas supplied to ordinary homes is so pure that the gas company deliberately adds mercaptan (S compound) in trace amounts to permit it to be detected by smell.

For at least the following reasons, the appellant requests review and reversal of this rejection.

The Appellant's Position

The comments above regarding the patentability of claim 1 in view of the same cited references apply equally here. Also, as indicated, claim 30 (and also claims 31, 32, and 33) are directed to products that can exploit the low ash content recited in parent claim 1 so as to yield, for example, enhanced organoleptics and CMA properties. As also indicated above, the Medalia article shows that conventional carbon blacks do not teach, suggest, or predict the success of such high purity carbon blacks as presently claimed in terms of the recited ash content and/or sulfur content. In particular, the Medalia article cited by the Examiner clearly and unequivocally teaches that a number of prior carbon blacks all had ash content *significantly exceeding* 0.1%. Medalia thus teaches away from the present claim 30. As also explained above, Medalia has not been shown by the Examiner to be "irrelevant" to the ash content expected of acetylene blacks of Yamazaki et al. Again, methane gas that is supplied to homes is not the same as industrial acetylene gas such as used by Yamazaki et al. Thus, the Examiner's example based on home gas at page 5 of the Final Office Action is inapposite.

Accordingly, for at least these above reasons, this rejection should be reversed.

Claim 31

Claim 31 recites an article that is a pressure pipe that comprises a polymer composition comprising the carbon black and a polymer of claims 1 and 4. The reasons for reversal as discussed above with respect to the rejection of parent claim 1 based on Yamazaki et al., Weaver et al., Dickerson, the Industrial Carbon reference, and the Medalia article, apply equally to claim 31.

Additional reasons for reversal are as follows.

A pipe, such as pressure pipe, is a recognized article having meaning to those skilled in the art with respect to shape, structure, and physical parameters. However, it is not a recognized "obvious expedient" to create a pipe from every polymer composition in existence, as suggested in the Final Office Action. The relied upon references do not provide an apparent reason why one of ordinary skill in the art would have made such a modification or why there would be a reasonable prediction of the success to achieve the results of the present invention.

Accordingly, this rejection should be reversed as well.

Claim 32

Claim 32 recites a pressure pipe that is a UV pressure pipe that comprises a polymer composition comprising the carbon black and a polymer of claims 1 and 4. The reasons for reversal as discussed above with respect to the rejection of parent claim 1 based on Yamazaki et al., Weaver et al., Dickerson, the Industrial Carbon reference, and the Medalia article, apply equally to claim 32. Additional reasons for reversal are as follows.

A pipe, such as a UV pressure pipe, is a recognized article having meaning to those skilled in the art with respect to shape, structure, and physical parameters. However, it is not a recognized "obvious expedient" to create a pipe from every polymer composition in existence, as suggested in the Final Office Action. The relied upon references do not provide an apparent reason why one of ordinary skill in the art would have made such a modification or why there would be a reasonable prediction of the success to achieve the results of the present invention.

Accordingly, this rejection should be reversed as well.

Claim 33

Claim 33 recites a potable water or gas pipe that comprises a polymer composition comprising the carbon black and a polymer of claims 1 and 4. The reasons for reversal as discussed above with respect to the rejection of parent claim 1 based on Yamazaki et al., Weaver et al., Dickerson, the Industrial Carbon reference, and the Medalia article, apply equally to claim 33. Additional reasons for reversal are as follows.

A pipe, such as a potable water or gas pipe, is a recognized article having meaning to those skilled in the art with respect to shape, structure, and physical parameters. However, it is not a recognized "obvious expedient" to create a pipe from every polymer composition in existence, as suggested in the Final Office Action. The relied upon references do not provide an apparent reason why one of ordinary skill in the art would have made such a modification or why there would be a reasonable prediction of the success to achieve the results of the present invention.

Accordingly, this rejection should be reversed as well.

7. **Rejection of Claims 1-3, 11-21, 24, 34-36, and 37-40 Under §103(a) Over Giet (U.S. Pat. No. 4,013,759) in view of Dickerson (U.S. Pat. No. 4,755,371), Weaver et al. (U.S. Pat. No. 5,352,289), the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)), and Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60).**

Claims 1-3, 11-21, 24, 34-36, and 37-40

Claims 1-3, 11-21, 24, 34-36, and 37-40 were finally rejected under 35 U.S.C. §102(b) as anticipated by or, in the alternative, under 35 U.S.C. §103(a) as obvious over U.S. Patent No. 4,013,759 to Giet in view of U.S. Patent No. 4,755,371 to Dickerson, U.S. Patent No. 5,352,289 to Weaver et al., the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of

Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)), and Industrial Carbon

(Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60).

The Examiner's Position

According to the Final Office Action (page 4), the Examiner interprets and applies Giet,

Dickerson, Weaver et al., the Medalia article, and Industrial Carbon, to claims 1-3, 11-21, 24, 34-

36, and 37-40 in the following manner:

> Giet teaches, in the entire patent but especially in column 6, high purity
> carbon black which can possess the claimed area due to the
> correspondence of iodine and nitrogen areas. It is noted that Giet does not
> actually specify which area is reported, so a difference may not exist. The
> 74 micron residue reported is 200 mesh; so the 200 mesh residue is known
> to be zero. Dickerson, Weaver, Medalia and Industrial Carbon are relied
> upon as above. Concerning claims 34-36, no difference is seen due to the
> similarity of the other properties.

For the following reasons, the appellant requests review and reversal of this rejection.

The Appellant's Position

The appellant observes that Giet describes only acetylene-like black having an electrical

resistivity of 0.4 to 0.7 ohm/cm (under pressure of 6.3 bars), a carbon content higher than 99%, an

adsorption capacity for water of filter index of at least about 80 gm of 3% acetonic water for 5 gm

of carbon black, a particle size less than about 200 A, a surface area in the range of 85 to 115

m^2/gm, and processes for making these acetylene-like blacks (see col. 8, lines 1-27).

Present claim 1 recites, *inter alia*, a 325 mesh (0.044 mm) residue of less than 20 ppm as

measured by ASTM D-1514. The appellant points out that the only apparent reference made in

Giet to a screen size is a mesh size of 0.074 mm, which the Examiner equates in the Final Office

Action with 200 mesh (see col. 6, lines 10-11). Further, this reference in Giet is made in the

limited context of a filter index measurement made on 5 g samples of black (see col. 6, lines 6-21).

In this respect, Giet indicates that a 5 g sample of black, on which the filter index value

measurement is conducted, is "previously screened through a screen having a mesh size of 0.074

mm" (col. 6, lines 10-11). Thus, for filter index testing, Giet teaches recovering the fraction of

black particles of the entire product sample that are smaller than about 0.074 mm before

conducting the filter index test on that smaller fraction. However, for the fraction of the black

particles that are 0.074 mm or larger, Giet provides no particle quantity information. Giet does not

state that the residue on the 0.074 mm (200 mesh) size screen was known to be zero, as alleged in

the Final Office Action. In fact, if there was zero residue on the 0.074 mm mesh size, there would

be logical reason for one skilled in the art to be conducting the sieving at that particular mesh size.

As also can be technically understood, particles large enough to be retained on 0.074 mm (200

mesh) size would inherently all be retained on an even smaller mesh size of 325 mesh (0.044 mm).

However, as indicated, Giet fails to provide the residue (retained) information for the larger 0.074

mm mesh size. Also, it can be technically appreciated that for the fraction of black particles of

Giet's black product that passes through 0.074 mm (200 mesh) size, it is theoretically possible that

none, some, or even all of that fraction could be retained as "residue" on a smaller mesh size of

325 mesh (0.044 mm). Thus, knowledge from Giet that some fraction of the black particles of an

acetylene black product passed through 0.074 mm (200 mesh) size does not permit any technical

inference to be drawn with any certainty about the 325 mesh residue quantity of that same product

sample.

With respect to particle sizes, Giet also indicates that the acetylene-like black has particle

sizes "in the range of 150 to 200 A ... measured by nitrogen adsorption" (col. 6, lines 4-48). The

appellant points out that particle size estimation by nitrogen adsorption does *not* correspond with

particle size measurements determined by ASTM D3849-89, such as recited for primary particle

sizes in present claim 1 on appeal. It is explained that primary particle size measurement according

to ASTM D3849-89 is a measurement of number-average primary particle size determined by

using a transmission electron microscope at a magnification of 30,000 for 100 particles selected at

random. For example, at col. 18, lines 23-27 of U.S. Patent No. 5,856,055, the patent explains

these details of the ASTM D3849-89 procedure. A partial copy of the above-referenced passages

in U.S. Patent No. 5,856,055, which was downloaded from the US PTO's website, is reproduced

in relevant part below.

5,856,055

17	18

1–70 nm, a BET specific surface area according to nitrogen adsorption (SBET) of at most 100 m²/g, a pH at least 2.0, a volatile matter content of at most 2 wt. %, and a DBP (dibutyl phthalate) absorptivity of 50–200 ml/g.
 	
  The carbon black may have an average primary article size of 10–70 nm, more preferably 20–60 nm, further preferably 25–45 nm. If the carbon black has an average primary particle size of below 10 nm, the polymerizable monomer composition containing the carbon black as well as the specific azo iron compound is liable to have an excessively large viscosity, and the fine particle formation thereof without agglomeration becomes difficult. Further, because of too small a primary particle size, it becomes difficult to disperse the carbon black in the polymerizable monomer composition. On the other hand, if the carbon black has an average primary particle size in excess of 70 nm, only a low coloring power can be attained even if it is well dispersed and, when used in a large amount in order to increase the coloring power, the resultant toner is caused to have a lower chargeability.
  It is preferred that the carbon black has a pH of at least 2.0, more preferably at least 4.0. If the carbon black is caused to have a pH below 2.0, the carbon black is caused to have many functional groups, which are liable to inhibit the polymerization.
  It is preferred that the carbon black has a specific surface area (S_BET) of at most 100 m²/g, more preferably 30–90 m²/g, further preferably 40–90 m²/g. The volatile matter content of the carbon black may preferably be at most 2 wt.
	compound and 10 wt. parts of carbon black was stirred by means of a stirrer at 200 rpm for 180 min. On the other hand, another polymerizable mixture was prepared similarly except for omitting the addition of the azo iron compound. The two mixtures were tested by standing for evaluation of dispersion stability. As a result, the mixture containing no azo iron compound caused precipitation after 3 days, whereas the mixture containing the azo iron compound was free from precipitation even after 30 days of standing.
	  The above-mentioned values for the respective properties of carbon black are based on values measured according to the follow ing methods:
	(1) DBP absorptivity
	  Measured by dripping DBP (dibutyl phthalate) onto a dry sample of carbon black (according to JIS K6221, A-method).
	(2) Specific surface area (S_BET) according to nitrogen adsorption
	  Measured according to ASTM D3037.
	(3) Volatile matter content
	  Determined by measuring the weights before and after heating of a carbon black sample (according to JIS K6221).
	(4) Average primary particle size
	  A number-average primary particle size is determined by using a transmission electron microscope at a magnification of 30,000 for 100 particles selected at random (according to ASTM D3849-89).
	(5) pH

   Thus, it is unknown and can not be predicted with certainty whether the primary particle

size values reported by Giet is equivalent to a primary particle size of not greater than 25 nm as

measured by ASTM D3849-89, as recited in claim 1 on appeal.

In addition, the Examiner equates the "surface area" values mentioned by Giet with the Iodine numbers recited in the present claims. First, Giet does not teach that the surface area values are nitrogen adsorption surface area values in particular, as assumed by the Examiner. Other possibilities exist, such as CTAB surface area values. Nor is any applicable surface area measurement standard indicated by Giet. Further, even assuming for sake of argument only, that Giet intended to refer to nitrogen adsorption surface area values, it is technically flawed to automatically assume that those values would numerically correspond with iodine absorption surface area values for the same product. With respect to the Examiner's assertion in the rejection that iodine numbers and nitrogen values have "correspondence," the appellant respectfully disagrees with this statement. If iodine numbers and nitrogen values correspond to each other, there would be no reason to have each test and yet the carbon black industry clearly measures carbon blacks using both tests as an ordinary practice in this field. Further, it is noted that the appellant previously submitted in the record before the Examiner a copy of an article on carbon black from the ENCYCLOPEDIA OF CHEMICAL TECHNOLOGY, Vol. 4, 4th Ed., 1992, with appellant's response of July 20, 2006, which summarizes the various test methods for surface area and other analytical methods. A copy of this article is included in the Evidence Appendix of this Appeal Brief. As shown in the included copy of the article, iodine adsorption is based on the amount of iodine adsorbed from an aqueous solution, wherein nitrogen surface area, such as BET surface area, is calculated from the amount of adsorbed nitrogen at liquid nitrogen temperature, (see Table 5, page 1058). Further, the supplied chart from the ENCYCLOPEDIA OF CHEMICAL TECHNOLOGY, namely Table 7 (see page 1061), further shows that iodine numbers can be different from nitrogen surface area. Accordingly, in view

of this evidence the Examiner's assumptions and justification for the rejection in this respect are not supported technically.

In the Final Office Action (page 2), the Examiner relies on the information provided on page 186 of the Dee Snell article for his proposition on the correlation or equivalence of nitrogen surface area and iodine numbers. The Examiner's reliance on the Dee Snell article as evidence showing iodine numbers and nitrogen surface areas correlate is flawed. The plot shown in Figure 7 on page 186 of Dee Snell may appear, for sake of argument, to generally show a correlation between nitrogen surface area and iodine adsorption for "furnace blacks." However, Figure 7 is explained in greater detail on subsequent page 187 of the Dee Snell article. The Dee Snell article clearly indicates on page 187 that the correlation for rubber grade carbons, such as shown in Figure 7, is not universal. In particular, the Dee Snell indicates that the iodine adsorption-nitrogen surface area correlation for rubber grade carbons may only apply to certain types of carbons, such as low-volatile, nonporous carbons. The Dee Snell article further explains on page 187 that "[d]eviations from linearity in the BET nitrogen-iodine adsorption correlation" occur with some types or classes of carbons, such as "carbons having a highly unsaturated oil on the surface or an oxygen volatile content per unit surface area significantly greater than the rubber grade furnace carbons." The Examiner has not identified any specific carbon black(s) in Giet that are considered by the Examiner to correspond to the specific types identified by Dee Snell that can have iodine and nitrogen surface area correlation, and as distinguished from other types of carbon blacks that would not correlate or be predicted to correlate according to the teachings of the Dee Snell article. Thus, the Dee Snell article is not evidence that shows with certainty that any carbon blacks of Giet inherently will be predicted and expected to have nitrogen surface area values that correlate with iodine adsorption values. Thus, insufficient

information is disclosed by Giet to permit or support an inference that the "surface area" values disclosed by Giet will correspond numerically to iodine values.

Giet is also silent on ash content and sulfur values of the acetylene-like blacks of that reference. Dickerson, Weaver et al., the Medalia article, and Industrial Carbon, do not cure the deficiencies of the primary reference to Giet, including in those respects, for similar reasons as explained above with respect to the rejection based on Yamazaki et al., and reference is made thereto.

Claims 2-3, 11-21, 24, 34-36, and 37-40, which ultimately depend from claim 1, are not anticipated by nor obvious over Giet, Dickerson, Weaver et al., the Medalia article, and Industrial Carbon, for at least the same reasons as set forth above relative to their parent claim 1.

In view of at least the above reasons, this rejection of claims 1-3, 11-21, 24, 34-36, and 37-40, on appeal based on Giet, Dickerson, Weaver et al., the Medalia article, and Industrial Carbon, should be reversed.

8. **Rejection of Claims 1-21, 24, 26-30, and 34-40 Under § 103(a) Over Giet (U.S. Pat. No. 4,013,759) in view of Dickerson (U.S. Pat. No. 4,755,371), Weaver et al. (U.S. Pat. No. 5,352,289), the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)), Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60), and van Konynenburg et al. (U.S. Patent No. 4,775,778)**

<u>Claims 1, 3-6, 10, 12-21, 24, 26, 28, 29, and 35-40</u>

Claims 1-21, 24, 26-30, and 34-40 were finally rejected under 35 U.S.C. §103(a) as obvious over U.S. Patent No. 4,013,759 to Giet in view of U.S. Patent No. 4,755,371 to Dickerson, U.S. Patent No. 5,352,289 to Weaver et al., the Medalia article (A.I. Medalia et al., "Particulate Carbon And Other Components Of Soot And Carbon Black," *Carbon*, Vol. 20, No. 6, pp.481-492 (1982)),

Industrial Carbon (Mantell, C.L., INDUSTRIAL CARBON, 2nd ed., 1946, p. 60), and U.S. Patent

No. 4,775,778 to van Konynenburg et al.

The Examiner's Position

According to the Final Office Action (page 4), the Examiner interprets and applies Giet,

Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al. to

claims 1-21, 24, 26-30, and 34-40 in the following manner:

> Giet not does [sic] show a polymer material, but teaches the advantages
> thereof in col. 1. Using the clamed [sic] polymers is an obvious expedient
> to provide an 'electro-conductive rubber' composition. The amount of
> carbon is deemed an obvious expedient of optimization of properties; the
> wide range of ratios include what appear to be conventional ratios- see
> Von Konynenburg [sic] col. 8 and 15. Note also In re Boesch 205 USPQ
> 215. Claim 30 appears to encompass normal shapes of extruded polymer.

For the following reasons, the appellant requests review and reversal of this rejection.

The Appellant's Position

Claim 1 on appeal recites an iodine number of from about 50 to about 112 mg/g, besides

other properties. Van Konynenburg et al. refers to "Regal 660" as Type No. 3 in Table 1 at column

13 of the reference. In the Office Action of April 20, 2006 (page 2), the Examiner specifically

identified and relied upon Regal 660 as a carbon black taught by van Konynenburg et al., which was

considered relevant by the Examiner. However, Regal 660 carbon black has an iodine number of

<u>125</u> mg/g. In this respect, the appellant directs attention to what is shown in a portion of U.S.

Patent No. 6,627,693 B1, and, particularly, see col. 5, Table 1, Sample 8 therein, which

corresponds to Regal 660. Also, at col. 5, line 5 U.S. Patent No. 6,627,693 B1, the table indicates

that ASTM procedure "D-1510" was used to measure iodine number for Sample 8, which is the

same ASTM standard test as used to measure the iodine number for purposes of the disclosure of

the present application (see page 17, line 18). A partial copy of the above-referenced passages in

U.S. Patent No. 6,627,693 B1, which was downloaded from the US PTO's website, is reproduced

in relevant part below.

US 6,627,693 B1

5

Property	ASTM Procedure
I₂No.	D-1510
CTAB	D-3765
Tint	D-3265
DBP	D-2414
N₂SA	D-3037

Table 1 sets forth the analytical properties of the carbon blacks of compounds 1–9. Compounds 2–9 contain furnace carbon blacks of the present invention. Compound 1 contains a conventional furnace carbon black control.

TABLE 1

	1 N650	2 A	3 B	4 C	5 D	6 E	7 F	8 G	9 H
I₂NO	36	82	121	258	258	258	356	125	150
DBP	122	102	114	117	105	64	133	60	143
Tint	56	103	115	148	153	151	141	138	146
CTAB	38	82	111	167	211	240	251	125	150
N₂SA	38	83	119	200	220	230	367	125	150

1 = STERLING VH; 2 = VULCAN 3; 3 = VULCAN 6; 4 = BP700; 5 = BP880, 6 = BP900; 7 = CRX1449; 8 = REGAL 660; and 9 = CRX1444, all tradenames of Cabot Corporation.

6

TABLE 2-continued

Ingredient	PHR
Wax	1.5
TBBS	1.5
Sulfur	1.25

The following test procedures were used to evaluate the physical properties of the rubber compositions of compounds 1–9:

Modulus, tensile, and elongation at break	D-412
Flex Fatigue-Cut Growth	MRRL Mk IV, Sinusoidal mode, tested at 43° C.
Abrasion Resistance	Akron Angle Abrader, 20° angle
Rebound	D-1054
Gloss	BYK-Gardner Micro TKI Gloss Meter
Jetness	Hunter Labscan 6000, 10°, D65 CIELAB Color Space

Most importantly, gloss was measured on tensile slabs not subjected to water using a BYK-Gardner Micro TKI Gloss meter under light reflecting at a 60° angle. Jetness was measured by the Hunter Labscan 6000, 10 degree, D65 CIELAB Color Space, measuring each compound versus a

As can be appreciated, the 125 mg/g iodine number of Regal 660 is numerically

significantly outside the about 50 to about 112 mg/g range recited for iodine number in claim 1 on

appeal. The appellant submits that the above-referenced portion of U.S. Patent No. 6,627,693 B1

is evidence that van Konynenburg et al. describes different carbon blacks from the present claims

with respect to at least one of the presently recited carbon black properties. Thus, the Examiner's

proposed modification of Giet in view of van Konynenburg et al. ignores important features of the

latter that would have lead one of ordinary skill in the art away from the present invention.

Further, it is respectfully noted that in van Konynenburg et al., no ASTM methods are

described. There is a reference to an article at col. 6, beginning at line 45, but no test procedures are identified. The test procedure used to measure various parameters can be quite relevant, since different test procedures can lead to different results. Thus, it is unclear with respect to van Konynenburg et al. whether a fair comparison can even be made with other parameters set forth in claim 1 on appeal, such as particle size. It is noted that in the present application, ASTM test numbers are provided for properties of iodine number, particle size, 325 mesh residue, ash content, sulfur content, CDBP and so forth (e.g., see p. 5, lines 12-17; page 6, line 21 to page 7, line 3; p. 17, lines 18-20, 23-24; and p. 18, lines 4-5) of the present specification. For example, for particle size, ASTM test procedure D3849-89 is used (see page 5, lines 12-13; page 18, lines 4-5). Moreover, when particle sizes on the order of nanometers are described, using a different test procedure can shift the measurement several nanometers and can be sufficient in being quite different from the ranges provided in the claimed invention. Further, van Konynenburg et al. does not teach or suggest any sulfur contents or ash contents, and did not consider it relevant from the standpoint of performance of the carbon black in polymer compositions

The appellant respectfully submits that there is no adequate factual basis that can be derived from van Konynenburg et al. to properly support and reach a conclusion that van Konynenburg et al. compensates for any of the above-identified deficiencies of Giet relative to claim 1 on appeal.

Further, Dickerson, Weaver et al., the Medalia article, and Industrial Carbon, do not cure above-indicated deficiencies of the primary reference to Giet relative to claim 1 for similar reasons as explained above with respect to the rejection based on Yamazaki et al., and reference is again made thereto.

Claims 2-21, 24, 26-30, and 34-40, which ultimately depend from claim 1, are not anticipated by nor obvious over Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al., for at least the same reasons as set forth above relative to their parent claim 1, and reference is made thereto. Furthermore, other than perhaps claims 7-9 (reciting carbon black amounts in the polymer compositions), the Final Office Action does not provide any analysis that articulates some reasoning or analysis with some rationale underpinning to support a legal conclusion of obviousness against dependent claims 2-21, 24, 26-30, and 34-40. This analysis should be made explicit. Therefore, there is insufficient analysis set forth by the Examiner in the Final Office Action to support a *prima facie* case of anticipation or obviousness against dependent claims 2-21, 24, 26-30, and 34-40 on appeal.

In view of the above, the appellant requests reversal of the rejection of claims 1-21, 24, 26-30, and 34-40 on appeal based on Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al.

Claim 2

Claim 2 further specifies that the I_2 No. is 73-104 mg/g. The reasons for reversal as discussed above with respect to the rejection of parent claim 1 based on Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al., apply equally to claim 2. Additional reasons for reversal are as follows.

The recitation of an Iodine number of 73-104 mg/g is even further distanced from the 125 mg/g iodine value applicable to Regal 660 in van Konynenburg et al., as explained above by the appellant. Carbon blacks of claim 2 on appeal are not obvious over Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and Konynenburg et al.

In view of the above, reversal of the rejection of claim 2 on appeal based on Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al. is respectfully requested.

Claims 7-9

Claims 7-9 recite a polymer composition comprising, per 100 parts by weight polymer, 0.5 to 300 parts by weight carbon black (claim 7), or 0.5 to 100 parts by weight carbon black (claim 8), or 0.5 to 80 parts by weight carbon black (claim 9). The reasons for reversal as discussed above with respect to the rejection of parent claim 1 based on Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al., apply equally to claims 7-9. Additional reasons for reversal are as follows.

Van Konynenburg et al. does not teach or suggest that the amount of acetylene-like blacks, such as those of Giet, as used in a polymer composition would have been an obvious expedient at the time of the present invention. Further, the Final Office Action does not indicate where van Konynenburg et al. teaches or suggests use of an acetylene black in any polymer composition. Further, the Examiner's reliance on the authority of *In re Boesch*, 205 USPQ 15, at page 4 of the Final Office Action is misplaced as van Konynenburg et al. does not teach or suggest at columns 8 and 15 thereof that the carbon black *amount or proportion* is a results-effective variable to be optimized (see M.P.E.P. §2144.05, citing *In re Boesch*).

In view of the above, reversal of the rejection of claims 7-9 on appeal based on Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al. is respectfully requested.

Claim 11

Claim 11 further specifies that the I_2 No. is 104 mg/g and the primary particle size is approximately 16 nm. The reasons for reversal as discussed above with respect to the rejection of parent claim 1 based on Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al., apply equally to claim 11. Additional reasons for reversal are as follows.

The recitation of I_2 No. is 104 mg/g and a primary particle size of approximately 16 nm is even further distanced from the 125 mg/g iodine value applicable to Regal 660 in van Konynenburg et al., as explained above. Van Konynenburg et al. also fail to teach or suggest or predict a carbon black combining the iodine and primary particle size values recited in claim 11 on appeal.

In view of the above, reversal of the rejection of claim 11 on appeal based on Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al. is respectfully requested.

Claim 27

Claim 27 recites a polymer comprising LLDPE, HDPE, MDPE, or combinations thereof. The reasons for reversal as discussed above with respect to the rejection of parent claim 1 based on Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al., apply equally to claim 27. Additional reasons for reversal are as follows.

Van Konynenburg et al. mentions fillers dispersed in *cross-linked* rubbers, elastomeric gums and thermoplastic elastomers (col. 3, line 63 to col. 4, line 10; col. 4, lines 56-59; col. 7, lines 23-26; col. 17, lines; abstract, lines 1-3), and not LLDPE, HDPE, MDPE, or combinations thereof. Further, claim 27 contains the carbon black of claim 1. As shown, van Konynenburg et al.

does not teach or suggest this carbon black. Therefore, Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon and van Konynenburg et al. does not render obvious claim 27 on appeal.

In view of the above, reversal of the rejection of claim 27 on appeal based on Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al. is respectfully requested.

Claim 30

Claim 30 recites an article that is a pipe, connector, cable jacketing, membrane, molding, or components thereof that comprise a polymer composition comprising the carbon black and a polymer of claims 1 and 4. The reasons for reversal as discussed above with respect to the rejection of claim 1 based on Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al., apply equally to claim 30. Additional reasons for reversal are as follows.

None of the relied upon references in the rejection show with evidence that claim 30 on appeal recites articles that "encompass normal shapes of extruded polymer," as suggested in the Final Office Action. The appellant further submits that it is not an "obvious expedient" to create the shaped polymeric articles of claim 30 from every polymer composition in existence.

Accordingly, this rejection should be reversed as well.

Claim 34

Claim 34, which depends on claim 1, recites a CDBP of less than or equal to 102 cc/100 g. The reasons for reversal as discussed above with respect to the rejection of parent claim 1 based on Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al., apply equally to claim 34. Additional reasons for reversal are as follows.

Van Konynenburg et al. states that the Regal 660 carbon black in Table 1 at column 13 having a DBP of 54 cc/100g must have nitrogen (N^2) absorption of 112 m^2/g. The van Konynenburg et al. reference does not suggest how one of ordinary skill can lower the nitrogen absorption of Regal 660 while maintaining the same indicated DBP property and primary particle size, nor suggest why such a modification might be desirable or provide predictable results.

In view of the above, reversal of the rejection of claim 34 on appeal based on Giet, Dickerson, Weaver et al., the Medalia article, Industrial Carbon, and van Konynenburg et al. is respectfully requested.

Conclusion

For the reasons set forth above, the appellant submits that the claims presently pending in the above-captioned application meet all of the requirements of patentability. It is therefore respectfully requested that the Honorable Board reverse the Examiner and remand this application for issue.

Respectfully submitted,

Luke A. Kilyk
Reg. No. 33,251

Atty. Docket No.: 02077 (3600-395-01)
KILYK & BOWERSOX, P.L.L.C.
400 Holiday Court, Suite 102
Warrenton, VA 20186
Tel.: (540) 428-1701
Fax: (540) 428-1720

(10)   Claims Appendix

1.   A carbon black having an I_2 No. of from about 50 to about 112 mg/g as measured by ASTM D1510, primary particle size of not greater than 25 nm as measured by ASTM D3849-89, and at least the following properties:

        a)   an ash content of less than 0.1% as measured by ASTM D-1506;

        b)   a total sulfur content of less than about 2% as measured by ASTM D-1619; and

        c)   a 325 mesh residue of less than 20 ppm as measured by ASTM D-1514.

2.   The carbon black of claim 1 wherein I_2 No. is 73-104 mg/g.

3.   The carbon black of claim 2 wherein the I_2 No. is 75-99 mg/g.

4.   A polymer composition comprising at least one polymer and the carbon black of claim 1.

5.   The polymer composition of claim 4 wherein the I_2 No. of the carbon black is 73-104 mg/g.

6.   The polymer composition of claim 4 wherein the I_2 No. of the carbon black is 75-99 mg/g.

7.   The polymer composition of claim 4 wherein the polymer composition comprises 0.5 to 300 parts by weight carbon black per 100 parts by weight of polymer.

8.   The polymer composition of claim 4 wherein the polymer composition comprises 0.5 to 100 parts by weight carbon black per 100 parts by weight of polymer.

9.   The polymer composition of claim 4 wherein the polymer composition comprises 0.5 to 80 parts by weight carbon black per 100 parts by weight of polymer.

10. The polymer composition of claim 4 wherein the polymer is a polyethylene or copolymers thereof.

11. The carbon black of claim 1 wherein the I_2 No. is approximately 104 mg/g and the primary particle size is approximately 16 nm.

12. The carbon black of claim 1 wherein the I_2 No. is approximately 89 mg/g and the primary particle size is approximately 18 nm.

13. The carbon black of claim 1 wherein the I_2 No. is approximately 91 mg/g and the primary particle size is approximately 18 nm.

14. The carbon black of claim 1 wherein the I_2 No. is approximately 99 mg/g and the primary particle size is approximately 17 nm.

15. The carbon black of claim 1 wherein the I_2 No. is approximately 86 mg/g and the primary particle size is approximately 19 nm.

16. The carbon black of claim 1 wherein the I_2 No. is approximately 96 mg/g and the primary particle size is approximately 17 nm.

17. The carbon black of claim 1 wherein the I_2 No. is approximately 85 mg/g and the primary particle size is approximately 17 nm.

18. The carbon black of claim 1 wherein the I_2 No. is approximately 73 mg/g and the primary particle size is approximately 18 nm.

19. The carbon black of claim 1 wherein the I_2 No. is approximately 86 mg/g and the primary particle size is approximately 19.5 nm.

20. The carbon black of claim 1 wherein the I_2 No. is approximately 90 mg/g and the primary particle size is approximately 19 nm.

21.   The carbon black of claim 1 wherein the I_2 No. is approximately 89 mg/g and the primary particle size is approximately 17 nm.

24.   The carbon black of claim 1, wherein said total sulfur content is less than about 0.1%.

26.   The polymer composition of claim 4, wherein said polymer is a polyolefin.

27.   The polymer composition of claim 4, wherein said polymer comprises LLDPE, HDPE, MDPE, or combinations thereof.

28.   The polymer composition of claim 4, wherein said polymer comprises a polystyrene, polycarbonate, nylon, or combinations thereof or copolymers thereof.

29.   An article comprising the polymer composition of claim 4.

30.   The article of claim 29, wherein said article is a pipe, connector, cable jacketing, membrane, molding, or components thereof.

31.   The article of claim 29, wherein said article is a pressure pipe.

32.   The article of claim 29, wherein said pressure pipe is a UV pressure pipe.

33.   The article of claim 29, wherein said article is a potable water or gas pipe.

34.   The carbon black of claim 1, further comprising a CDBP of less than or equal to 102 cc/100 g, as measured by ASTM D3493-86.

35.   The carbon black of claim 34 wherein the CDBP is 70-100 cc/100 g.

36.   The carbon black of claim 34 wherein the CDBP is 80-95 cc/100 g.

37.   The carbon black of claim 1, having an I_2 No. of 50-85 mg/g; a primary particle size of less than or equal to 25 nm; and a CDBP of less than or equal to 96 cc/100 g, as measured by ASTM D3493-86.

38.   The carbon black of claim 37 wherein the I_2 No. is 55-80 mg/g.

39.   The carbon black of claim 37 wherein the primary particle size is from greater than 20 nm to 25 nm.

40.   The carbon black of claim 37 wherein the CDBP is 50-96 cc/100 g.

41.   The carbon black of claim 32 wherein the I_2 No. is 60-78 mg/g.

(11)   Evidence Appendix

      (I)   ENCYCLOPEDIA OF CHEMICAL TECHNOLOGY,
                "Carbon (Carbon Black)", E.M. Dannenberg,
                Vol. 4, 4th Ed., 1992, pp. 1037-1074,
                New York, Wiley-Interscience (39 pages). [2]

      (II)   ENCYCLOPEDIA OF INDUSTRIAL CHEMICAL ANALYSIS
                Ed. by F. Dee Snell et al., Vol. 8, pp. 163, 186-187, 243, *Interscience Publishers*,
                (6 pages).[3]

---

[2] Cited with the Appellants' Amendment filed July 20, 2006.
[3] Cited by Appellant on form PTO/SB/08a filed July 20, 2006, as part of a "Non Patent Literature Documents" listing described as "Communication of a Notice of Opposition from European Patent Office dated June 6, 2005 and English translation of Opposition filed by Degussa AG on May 10, 2005 in European Patent No. 1 114 848," which submission included a copy of the Dee Snell document, which was identified on the leading page as opposition reference document "D10."

EVIDENCE APPENDIX I

CARBON (CARBON BLACK)

E. M. DANNENBERG

CARBON BLACK

Carbon black is a generic term for an important family of products used principally for the reinforcement of rubber, as a black pigment, and for its electrically conductive properties. It is a fluffy powder of extreme fineness and high surface area, composed essentially of elemental carbon. Plants for the manufacture of carbon black are strategically located worldwide in order to supply the rubber tire industry, which consumes 70% of production. About 20% is used for other rubber products and 10% is used for special nonrubber applications. World capacity in 1988 was estimated at over six million metric tons; U.S. capacity was almost 1.6 million metric tons. Carbon black was ranked 38 in 1989 among the 50 largest volume industrial chemicals produced in the United States. Six U.S. manufacturers (1) were operating 22 plants in 1990. Many of these are located in the south and southwest. Over 35 grades, listed in ASTM 1765-87 (2), are used by the rubber industry, and one manufacturer supplies an additional 45 grades for special pigment applications.

Carbon blacks differ from other forms of bulk carbon such as diamond, graphite, cokes, and charcoal in that they are particulate, composed of aggregates having complex configurations, quasigraphitic in structure, and of colloidal dimensions. They differ from other bulk carbons in having their origin in the vapor phase through the thermal decomposition and the partial combustion of hydrocarbons. Carbon black is a product of a process incorporating the latest engineering technology and process controls. Its purity differentiates it from soots that are impure by-products from the combustion of coal and oils and from the use of diesel fuels. Carbon blacks are essentially free of the inorganic contaminants and extractable organic residues characteristic of most forms of soot.

A number of processes have been used to produce carbon black including the oil-furnace, impingement (channel), lampblack, and the thermal decomposition of natural gas and acetylene (3). These processes produce different grades of carbon

and are referred to by the process by which they are made, eg, oil-furnace black, lampblack, thermal black, acetylene black, and channel-type impingement black. A small amount of by-product carbon from the manufacture of synthesis gas from liquid hydrocarbons has found applications in electrically conductive compositions. The different grades from the various processes have certain unique characteristics, but it is now possible to produce reasonable approximations of most of these grades by the oil-furnace process. Since over 95% of the total output of carbon black is produced by the oil-furnace process, this article emphasizes this process.

History of Carbon Black Manufacture

The use of carbon black as a pigment dates back to prehistoric times. Cave wall dwellings and objects from ancient Egypt were decorated with paints and lacquers containing carbon black. The oldest process practiced in China about 3000 BC consisted of the partial combustion of vegetable oils in small lamps with ceramic covers. The smoke impinged on the covers from which the adhering carbon black was carefully removed. Another old process is the lampblack process, which is the ancestor of all modern carbon blacks. Until the 1870s it was the only commercial process, and because of this the word lampblack is occasionally used as a generic term for carbon black. In the lampblack process, oils are burned in open, shallow pans in a restricted air supply. The heavy, carbon-laden smoke is passed through a series of settling chambers and filters from which the flocculated carbon deposits are recovered.

Prior to 1870 it was already known that carbon black with much higher covering power and jetness could be recovered from underventilated illuminating gas flames impinging on a cold surface. These gas blacks led to the development of the channel process, the name deriving from the iron channels used for the collection of the carbon blacks from the impingement of thousands of small luminous flames burning in a restricted atmosphere of air. This process dominated the industry for over 50 years. In 1926 there were 33 producers in the United States. Because of poor carbon yields from natural gas in the range of 1–5% and severe atmospheric pollution, this process has become extinct. The last channel black plant in the United States was closed in 1976.

In the 1920s two other processes using natural gas were introduced that gave much higher yields with large decreases in atmospheric contamination. One was the cyclic thermal black process. Alternate heating and production cycles in large brick checkered chambers are used to produce a unique large particle size, essentially unaggregated-grade useful for many special rubber and plastic applications. Thermal black is produced in the United States, Canada, England, and a few other locations worldwide. The other process, based on natural gas, was the so-called gas-furnace process and is no longer used. This process was continuous and the forerunner of the oil-furnace process. It was discontinued because of the relatively low yield, high raw material cost, and limited range of products.

The first commercial oil-furnace process was put into operation in 1943 by the Phillips Petroleum Co. in Borger, Texas. The oil-furnace blacks rapidly displaced all other types used for the reinforcement of rubber and today account

for practically all carbon black production. In the oil-furnace process heavy aromatic residual oils are atomized into a primary combustion flame where the excess oxygen in the primary zone burns a portion of the residual oil to maintain flame temperatures, and the remaining oil is thermally decomposed into carbon and hydrogen. Yields in this process are in the range of 35 to 50% based on the total carbon input. A broad range of product qualities can be produced.

Before World War I carbon black was almost exclusively used as a black pigment for printing inks, paints, and enamels. The singular event that changed the industry from a small specialty manufacturer to large volume producer of a vital raw material was the discovery of rubber reinforcement in 1904 (4). The automobile and the tire industries were expanding rapidly, and there was a demand for longer wearing automobile tires. The use of carbon black as a filler for rubber fulfilled this need providing longer wearing and more durable pneumatic tires. The use of carbon black in tires remains its most important application, coupling the fortunes of the carbon black industry to that of the automotive industry.

Physical Structure of Carbon Black

Molecular and Crystallite Structure. The arrangement of carbon atoms in carbon black has been well-established by x-ray diffraction methods (5,6). The diffraction patterns show diffuse rings at the same positions as diffraction rings from pure graphite. The suggested relation to graphite is further emphasized as carbon black is heated to 3000°C. The diffuse reflections sharpen, but the pattern never achieves that of true graphite. Carbon black can have a degenerated graphitic crystallite structure. Whereas graphite has three-dimensional order, as seen in the model structures of Figure 1, carbon black has two-dimensional order. The x-ray data indicate that carbon black consists of well-developed graphite platelets stacked roughly parallel to one another but random in orientation with respect to adjacent layers. As shown in Figure 1 the carbon atoms in the graphite structure form large sheets of condensed aromatic ring systems with an interatomic spacing of 0.142 nm, comparable to the aromatic carbon separation distance of 0.139 nm in benzene. The large graphite interplanar distance of 0.335 nm results in a specific gravity of 2.26. In carbon black the interplanar distance is still larger, in the range of 0.350–0.365 nm, as a consequence of the random planar orientations or so-called turbostratic arrangement. The specific gravities of commercial carbon blacks are 1.76–1.90 depending on the grade. X-ray diffraction data provide estimates of crystallite size. L_a is the average layer plane diameter and L_c is the average crystallite thickness. For a typical carbon black L_a is 1.7 nm and L_c is 1.5 nm, which corresponds to an average of four layer planes per crystallite containing 375 carbon atoms. A particle of a 100 m^2/g carbon black contains over 4000 crystallites. It was originally suggested that these discrete crystallites were in random orientation within the particle. This view was later abandoned when electron microscopy of graphitized and oxidized carbon blacks indicated more of a concentric layer plane arrangement. This structure has been confirmed by the use of high resolution phase-contrast electron microscopy that made possible the direct imaging of graphitic layer planes in carbon black (7).

Figure 2 shows a phase-contrast electron micrograph of carbon black at high resolution that displays the marked concentric arrangement of the layer planes at the surface and around what appear to be growth centers.
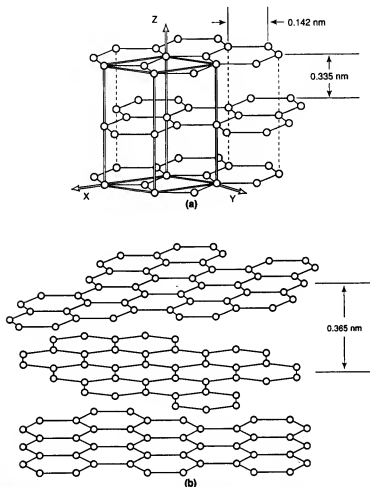

Fig. 1.   Atomic structural models of (a), graphite, and (b), carbon black.

The word particle has become so widely used in the technical rubber and carbon black literature that it is convenient to retain the term when in fact nodule is meant. The layer planes are curved, distorted, and of varying size. They also intersect and interconnect one particle or nodule with its neighbors. This type of structure has been termed paracrystalline. It is obvious that individual particles do not exist in carbon blacks, with the exception of thermal blacks, and that the functional unit is an aggregate of nodules that probably existed as smaller particles at some early stage of the carbon formation process. The functional unit in well-dispersed systems is called an aggregate.
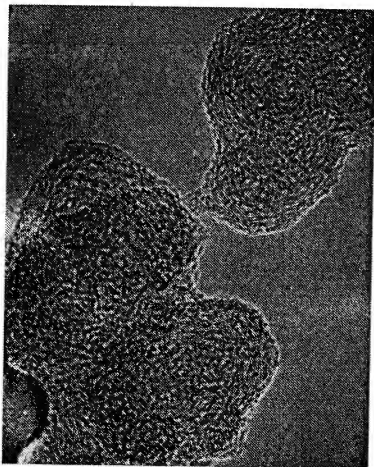
Fig. 2. High resolution (3,000,000 ×) electron micrograph of H-300-grade carbon black. Courtesy of W. M. Hess.

Morphology. In describing carbon black, three terms are used to describe structures of increasing scale and complexity:

Particles (nodules) are the primary structure element. They are roughly spherical elements that are joined in the aggregate structures.

Aggregates are the primary dispersable elements of carbon black in all but thermal blacks. The particles in an aggregate are connected and have grown together.

Agglomerates are undispersed clusters of aggregates held together by van der Waals forces or by binders. The term structure is used to describe both the extent and the complexity with which the particles are interconnected in aggregates. Primary measures of structure focus on the internal space within the aggregate.

Size and shape of the aggregates in composite systems are the principal features that determine the performance of carbon black as a reinforcing agent and as a pigment (8). Figure 3 shows an electron micrograph of a reinforcing tread black. There is an enormous range in aggregate size. The aggregate size distribution curve for N220 shown in Figure 4 is log-normal, and the range of D_c, equivalent diameters of the projected areas of the aggregates is about tenfold. Within each aggregate the nodules, or particles, appear to be about the same size. The size of the aggregates is directly related to the size of the particles. The shapes of the aggregates have infinite variety from tight grapelike clusters to open dendritic or branched arrangements to fibrous configurations.
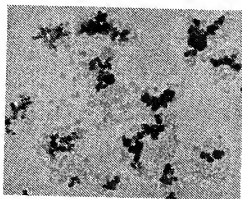

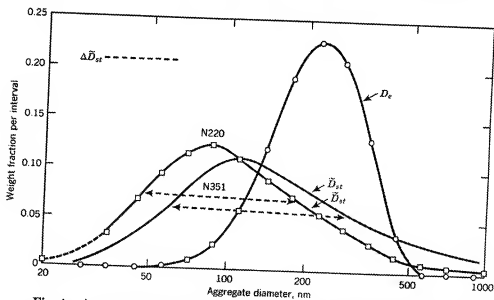**Fig. 3.**   Electron micrograph of reinforcing-grade of N399 tread black (100,000 ×).

Fig. 4.   Aggregate size distributions by electron microscope image analysis (D_e) and centrifugal ($\bar{D}_{St}$) sedimentations for N220 and N351 carbon blacks (8).

A useful method for determining relative aggregate sizes and distributions is by centrifugal sedimentation. From the sedimentation rates of the aggregates the Stokes diameter is derived. A convenient instrument for these measurements is the Joyce Loebl disk photosedimentometer (9). Large aggregates sediment at a faster rate than smaller ones. The sedimentation rate is also influenced by the bulkiness of the aggregates. At constant volume or mass, a bulky aggregate sediments more slowly than a compact aggregate because of frictional drag. Figure 4 (8) shows a comparison of a Stokes diameter distribution $\tilde{D}_{St}$ and equivalent diameter distribution D_e from electron microscopy for N220. In this example the modal $\tilde{D}_{St}$ value is about one-third of the modal D_e value.

Table 1 lists average $\tilde{D}_{St}$ values from a number of literature sources. This table also lists d_{um} values for the aggregates calculated from their estimated volumes. In this case there is reasonable agreement between the two diameters. Aggregate size distributions from centrifugal sedimentation analysis are very useful for assessing the differences in this characteristic within a given grade or at constant surface area. It has been shown that the hysteresis of rubber vulcanizates can be reduced by broadening the aggregate size distribution curve without any significant loss in abrasion resistance (11,12). As shown in Figure 4 this broadening is usually expressed as ΔD_{50}, the width at 50% of the modal value. $\tilde{D}_{St}$ values have been related to the dynamic and mechanical performance of rubber-grade carbon blacks. Hysteresis decreases and abrasion loss increases with increasing values of $\tilde{D}_{St}$ (13).

Table 1. Carbon Black Morphology[a]

ASTM designation	Particle size, d_{um}[b], nm	Aggregate size, d_{um}[b], nm	$\tilde{D}_{St}$[c], nm	Surface area, m²/g
N110	27	93	76–111	143
N220	32	103	95–117	117
N234	31	109	74–97	120
N326	41	108	98	94
N330	46	146	116–145	80
N339	39	122	96–125	96
N351	50	159	127	75
N375	36	106	91	105
N550	93	240	220–242	41
N660	109	252	227–283	34
N774	124	265	261	30
N990	403	593	436	9

[a]Ref. 10. Particle size, aggregate size, and surface area are by em.

[b]d_{um} = weight mean diameter = $\dfrac{nd^4}{nd^3}$.

[c]Stokes diameter by centrifugal sedimentation from various sources.

The tinting strength of rubber-grade carbon blacks shows a linear relationship with $\tilde{D}_{St}$ shown in Figure 5. Since performance characteristics are known to

depend on aggregate volume, surface area, and bulkiness, it appears that the $\bar{D}_{St}$ values combine the effects of all these factors. As such, it is a valuable addition to carbon black characterization methodology.
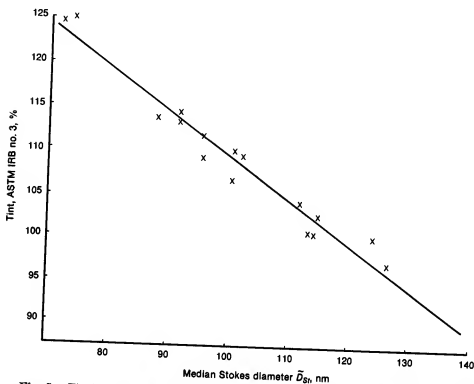

Fig. 5. Tinting strength versus median Stokes diameters for a range of reinforcing tread blacks.

Aggregate Morphology and Structure. The term structure is widely used in the carbon black and rubber industries. It was originally introduced in 1944 (14) to describe a chaining tendency of the carbon black particles. It is now used to describe the relative void volume characteristics of grades of black of the same surface area. Structure comparisons of grades with different surface areas cannot be made. It is now known that the properties associated with structure are associated principally with the bulkiness of individual aggregates. Aggregates of the same volume, surface area, and number of nodules have high structure in an open bulky and filamentous arrangement and a low structure in a more clustered compact arrangement.

High structure blacks in unvulcanized rubber give higher Mooney viscosities, lower die swell, faster extrusion rates, and better and more rapid dispersion after incorporation. In vulcanized rubber higher modulus is obtained. High structure blacks give lower bulk densities and high vehicle demand in paint systems.

Structure is usually measured by a void volume test such as the absorption

of dibutyl phthalate (DBPA) (15), or by bulk density measurements of the carbon black under compression. In order to eliminate the effects of pelletizing conditions the DBPA test has been modified to use a sample that has been precompressed at a pressure of 165 MPa (24,000 psi) and then broken up four successive times (24M4) (16). This procedure causes some aggregate breakdown and is claimed to more closely approximate the actual breakdown that occurs during rubber mixing.

Aggregate Breakdown. Aggregate size analysis by the electron microscope and centrifuge methods are performed on predispersed samples of carbon black. High shear energy, usually ultrasonic, and enough time are employed in these sample preparations to break down microagglomerates to their ultimate, aggregates for measurement. When mixed into elastomers under high shear conditions the aggregates themselves undergo fracture forming smaller aggregates that become the actual functional units (17–19). The extent of breakdown depends on shearing stress, energy input, and the grade of carbon black. Elastomer mixes were studied using the techniques of ultramicrotome and automated image analysis. Ultrasonic dispersions of carbon gel preparations from elastomer mixes have also been used in breakdown studies. A high DBPA reinforcing tread grade (N347) exhibited a significant reduction in aggregate length in a BR/OEP tread formulation, whereas a low DBPA grade (N326) showed no measurable change. The extent of aggregate length reduction was 30 to 40% for the normal and high DBPA grades (20).

The effect of elastomer viscosity on aggregate breakdown has been shown (19). A high DBPA grade (N339) was well-mixed with a 52 and a 100 Mooney viscosity OE-SBR. A 43% reduction in aggregate volume was reported for the 52 Mooney rubber and a 53% reduction for 100 Mooney rubber. High resolution electron micrographs show actual fracture locations at the ends of aggregates. The extent of fracture from aggregate length and volume breakdown is consistent with one average fracture per aggregate for the high DBPA grades.

Chemical Composition

Oil-furnace blacks used by the rubber industry contain over 97% elemental carbon. Thermal and acetylene black consist of over 99% carbon. The ultimate analysis of rubber-grade blacks is shown in Table 2. The elements other than carbon in furnace black are hydrogen, oxygen, and sulfur, and there are mineral oxides and salts and traces of adsorbed hydrocarbons. The oxygen content is located on the surface of the aggregates as C_xO_y complexes. The hydrogen and sulfur are distributed on the surface and the interior of the aggregates. Some special blacks used for pigment purposes contain larger quantities of oxygen than normal furnace blacks. These blacks are made by oxidation in a separate process step using nitric acid, ozone, air, and other oxidizing agents. They may contain from 2 to 6% oxygen. Oxidation improves dispersion and flow characteristics in pigment vehicle systems such as lithographic inks, paints, and enamels. In rubber-grade blacks surface oxidation reduces pH and changes the kinetics of vulcanization, making the rubber compounds less scorchy and slower curing.

Table 2. Chemical Composition of Carbon Blacks, %

Type	Carbon	Hydrogen	Oxygen	Sulfur	Ash	Volatile
rubber-grade furnace	97.3–99.3	0.20–0.40	0.20–1.20	0.20–1.20	0.10–1.00	0.60–1.50
medium thermal	99.4	0.30–0.50	0.00–0.12	0.00–0.25	0.20–0.38	
acetylene	99.8	0.05–0.10	0.10–0.15	0.02–0.05	0.00	<0.40

A convenient method for assessing the extent of surface oxidation is the measurement of volatile content. This standard method measures the weight loss of the evolved gases on heating up to 950°C in an inert atmosphere. The composition of these gases consists of three principal components: hydrogen, carbon monoxide, and carbon dioxide. The volatile content of normal furnace blacks is under 1.5%, and the volatile content of oxidized special grades is 2.0 to 9.5%.

The origin of the volatile gases is the functional groups attached to the carbon black layer planes. These groups are carbon-bound hydrogen, phenols, hydroquinones, quinones, neutral groups with one oxygen, carboxylic acids, lactones, and neutral groups containing two oxygens (21). Hydrogen is the most dominant of these groups. The oxygen content is present mainly as weakly acidic phenolic groups located at the surface of the aggregates. Figure 6 shows an idealized graphite surface layer plane with the various functional groups located at the periphery of the plane.
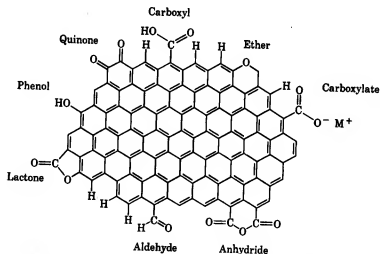
Fig. 6.   Aromatic layer plane with functional side groups.

In addition to combined hydrogen and oxygen, carbon blacks may contain as much as 1.2% combined sulfur resulting from the sulfur content of the aromatic feedstock that contains thiophenes, mercaptans, and sulfides. The combined sulfur appears to be inert and does not contribute to sulfur cross-linking during the vulcanization of rubber compounds.

	Volatile
0	0.60–1.50
8	<0.40

ation is the
weight loss
he composi-
gen, carbon
ce blacks is
0 to 9.5%.
ched to the
n, phenols,
xylic acids,
is the most
eakly acidic
6 shows an
ups located

te

r contain as
ne aromatic
nbined sul-
during the

The ash content of furnace blacks is normally a few tenths of a percent but in some products may be as high as one percent. The chief sources of ash are the water used to quench the hot black from the reactors during manufacture and for wet pelletizing the black. The hardness of the water, and the amount used determines the ash content of the products. The ash consists principally of the salts and oxides of calcium, magnesium, and sodium and accounts for the basic pH (8–10) commonly found in furnace blacks. In some products potassium, in small amounts, is present in the ash content. Potassium salts are used in most carbon black manufacture to control structure and rubber vulcanizate modulus (22). The basic mineral salts and oxides have a slight accelerating effect on the vulcanization reaction in rubber.

Carbon Black Formation Mechanisms

The formation of carbon black in a candle flame was the subject of a series of lectures in the 1860s by Michael Faraday at the Royal Institution in London (23). Faraday described the nature of the diffusion flame, the products of combustion, the decomposition of the paraffin wax to form hydrogen and carbon, the luminosity of the flame because of incandescent carbon particles, and the destructive oxidation of the carbon by the air surrounding the flame. Since Faraday's time, many theories have been proposed to account for carbon formation in a diffusion flame, but controversy still exists regarding the mechanism (24).

Mechanisms of formation must account for the unique morphology and microstructure of carbon black. These features include the presence of nodules, or particles, multiple growth centers within some nodules, the fusion of nodules into large aggregates, and the paracrystalline or concentric layer plane structure of the aggregates. One mechanism of formation involves the decomposition of the aromatic hydrocarbon fuel in a diffusion flame to hydrogen and carbon radicals, and carbon–hydrogen radical fragments. These combine into larger aromatic layer plane units until they are no longer stable and condense out of the vapor phase to form nuclei, or growth centers. Further carbon deposition forms carbon particles that are the precursors of the nodules. The carbon particles collide and coalesce while undergoing further deposition of carbon layer planes and their surface, forming the nodules and aggregates with their characteristic onion microstructure as seen in the micrographs (25). The various steps in the sequence are not well understood. There is particular disagreement regarding the nucleation and particle formation steps preceding the formation of nodules. One suggestion is that the particles go through a fairly sticky stage as they collide and coalesce to form the aggregates. Another suggestion is that the layer planes formed in the vapor phase condense out to form solid multiple layer plane nuclei. Carbon deposition on the nuclei results in particles and eventually nodules and aggregates. The remarkable and industrially important influence of ionic species such as K^+ and Ca^{2+} on the morphology of the aggregates and their surface area during the carbon black formation process is a strong indication that ionic mechanisms may be active in the nucleation and aggregate formation steps (22). There are several reviews of carbon formation mechanisms (26,27).

Manufacture

THE OIL-FURNACE PROCESS

The oil-furnace process, based on the partial combustion of liquid aromatic residual hydrocarbons, was first introduced in the United States at the end of World War II. It rapidly displaced the then dominant channel (impingement) and gas-furnace processes because it gave improved yields and better product qualities. It was also independent of the geographical source of raw materials, a limitation on the channel process and other processes dependent on natural gas, making possible the worldwide location of manufacturing closer to the tire customers. Environmentally it favored elimination of particulate air pollution and was more versatile than all other competing processes.

A simplified flow diagram of a modern furnace black production line is shown in Figure 7 (28). The principal pieces of equipment are the air blower, process air and oil preheaters, reactors, quench tower, bag filter, pelletizer, and rotary dryer. The basic process consists of atomizing the feedstock into the combustion zone of the reactor where the combustion of natural gas and preheated excess air create a high temperature environment of 1200 to 1900°C that almost instantly vaporizes the feedstock and decomposes most of it to carbon black and hydrogen. The remaining feedstock reacts with the excess oxygen in the primary combustion stream to maintain the reaction temperature for carbon formation. In some reactors a number of feedstock streams are atomized radially into the high velocity combustion gases. The reaction products must be quenched rapidly with water sprays to lower the temperature to prevent loss of the carbon black product through reaction with carbon dioxide and water, products of the combustion reactions. The hot, heavy carbon black smoke from the reactors enters the air preheater where thermal energy is transferred to preheat the primary combustion air. From the air preheater the lower temperature combustion products are given a secondary quench for a further lowering of temperature in a tower from which they enter the bag filter that separates the fluffy carbon black product from the tail gases. Since the tail gases are composed mainly of water, nitrogen, carbon monoxide, carbon dioxide, and hydrogen, they have heating value as a fuel to supplement the natural gas used to preheat feedstock and for heating the pellet dryers. Unused tail gas is frequently flared prior to venting to the atmosphere after removal of particulate matter. The fluffy carbon black from the bag filter is mechanically agitated to increase its bulk density and is then conveyed to the wet pelletizers where water is added to transform the product into wet granules. Dry pelletization in rotating drums is practiced for some special applications. The wet pellets are then dried in a rotary dryer after which finished product goes to storage tanks for shipping in bulk or in bags.

Feedstocks. Feedstocks are viscous aromatic hydrocarbons consisting of branched polynuclear aromatics with smaller quantities of paraffins and unsaturates. Preferred feedstocks are high in aromaticity, free of coke and other gritty materials, and contain low concentrations of asphaltenes, sulfur, and alkali metals. Other limitations are the quantities available on a long-term basis, uniformity, ease of transportation, and cost. The ability to handle such oils in tanks, pumps, transfer lines, and spray nozzles are also primary requirements.
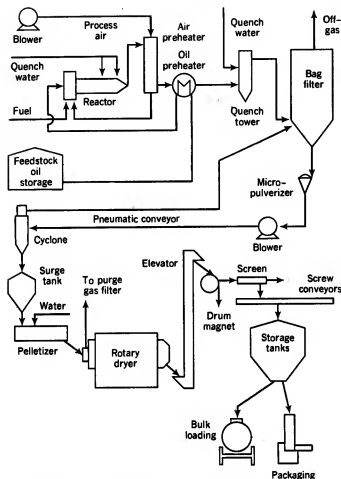
Fig. 7. Flow diagram of oil-furnace black process.

The principal sources of feedstocks in the United States are the decant oils from petroleum refining operations. These are clarified heavy distillates from the catalytic cracking of gas oils. About 95% of U.S. feedstock use is decant oil. Another source of feedstock is ethylene process tars obtained as the heavy by-products from the production of ethylene by steam cracking of alkanes, naphthas, and gas oils. There is a wide use of these feedstocks in European production. European and Asian operations also use significant quantities of coal tars, creosote oils, and anthracene oils, the distillates from the high temperature coking of coal. European feedstock sources are 50% decant oils and 50% ethylene tars and creosote oils.

Aromaticity is the most important property of a carbon black feedstock. It is generally measured by the Bureau of Mines Correlation Index (BMCI) and is an indication of the carbon-to-hydrogen ratio. The sulfur content is limited to reduce corrosion, loss of yield, and sulfur in the product. It may be limited in certain locations for environmental reasons. The boiling range must be low enough so that it will be completely volatilized under furnace time–temperature conditions.

Alkane insolubles or asphaltenes must be kept below critical levels in order to maintain product quality. Excessive asphaltene content results in a loss of reinforcement and poor treadwear in tire applications.

The pricing of carbon black feedstocks depends on their alternate market as residual fuel oil, especially that of high sulfur No. 6 fuel oil. The actual price is determined by the supply/demand relationships for these two markets. Feedstock cost contributes about 60% of the total manufacturing cost. The market price of carbon black is strongly dependent on the feedstock cost as shown in Figure 8.
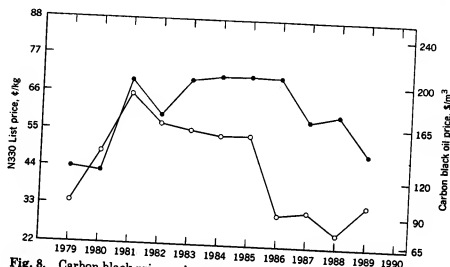

Fig. 8. Carbon black price and raw material cost in the United States (1979–1989). Average U.S. list price N330 (HAF)-grade carbon black, ¢/kg. To convert $/m^3 to $/barrel, multiply by 0.159.

Reactors. The heart of a furnace black plant is the furnace or reactor where carbon black formation takes place under high temperature, partial combustion conditions. The reactors are designed and constructed to be as trouble-free as possible over long periods of operation under extremely aggressive conditions. They are monitored constantly for signs of deterioration in order to ensure constant product quality. The wide variety of furnace black grades for rubber and pigment applications requires different reactor designs and sizes to cover the complete range, though closely related grades can be made in the same reactor by adjusting input variables. Reactors for higher surface area and reinforcing grades operate under high gas velocities, temperatures, and turbulence to ensure rapid mixing of reactant gases and feedstock. Lower surface area and less reinforcing grades are produced in larger reactors at lower temperatures, lower velocities, and longer residence time. Table 3 lists carbon formation temperatures, residence times, and maximum velocities for the complete surface area range of rubber-grade blacks. The N-series designation is in accordance with ASTM D1765, which is the standard classification system for carbon blacks used in rubber products (15). At least three different reactor designs must be used to make this range of furnace blacks and thermal black.

Table 3. Time–Temperature–Velocity Conditions in Carbon Black Reactors[a]

	Surface area, m^2/g	Temperature, °C	Residence time, s	Maximum velocity, m/s
N100 series, SAF[b]	145	1800	0.008	
N200 series, ISAF[b]	120		0.010	180–400
N300 series, HAF[c]	80	1550	0.031	
N500 series, FEF[d]	42		1.0	30–80
N700 series, SRF[e]	25	1400	1.5	0.5–1.5
N990 thermal	8	1200–1350	10	10

[a]These characteristic conditions and values depend on reactor designs and fuel rates.
[b]SAF = super abrasion furnace; ISAF = intermediate super abrasion furnace.
[c]HAF = high abrasion furnace.
[d]FEF = fast extrusion furnace.
[e]SRF = semireinforcing furnace.

Reactors are built to have three fairly well-defined zones. Gas and air are introduced into an upstream, primary combustion zone. For reinforcing grades, this connects with a mixing zone of high velocity and turbulence where feedstock is introduced as a fine atomized spray. The mixing zone is followed by a reaction zone of cylindrical shape where carbon-forming reactions occur. Downstream of the reaction zone is a water quench. For high surface area blacks the reactors may have a 15 to 38 cm diameter mixing zone with lengths up to five m. For lower area blacks the reactors are cylindrical with diameters of 75 cm or more and lengths of 9 to 12 m. There is a wide variety of reactors, and each manufacturer has proprietary designs. Air and gas may be introduced to the primary combustion zone either axially, tangentially, or radially. The feedstock can be introduced into the primary flame either axially or radially in the high velocity section of the mixing zone. The high velocity section may be venturi-shaped or consist of a narrow diameter choke. The reactors have a steel shell construction lined with high temperature-resistant castable refractories and insulating cements. The refractories have a service life of one to three years. Figures 9 and 10 show the designs of commercial reactors based on the patent literature.

The quality and yield of carbon black depends on the quality of the feedstock, reactor design, and input variables. The structure is controlled by the addition of alkali metals to the reaction or mixing zones. Usual practice is to use aqueous solutions of alkali metal salts such as potassium chloride or potassium hydroxide sprayed into the combustion chamber or added to the make oil in the oil injector. Alkaline-earth compounds such as calcium acetate that increase the specific surface area are introduced in a similar manner.

The energy utilization in the production of one kilogram of oil-furnace carbon black is in the range of $9.3–16 \times 10^7$ J (4–6.9 × 10^4 Btu/lb), and the yields are 300–660 kg/m^3 (2.5–5.5 lb/gal) depending on the grade. The energy inputs to the reactor are the heat of combustion of the preheated feedstock, heat of combustion of natural gas, and the thermal energy of the preheated air. The energy output consists of the heat of combustion of the carbon black product, the heat of combustion and the sensible heat of the tail gas, the heat loss from the water
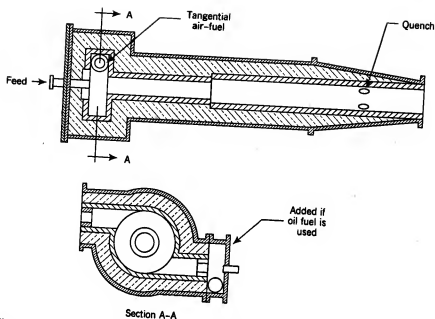
Fig. 9. Reactor for HAF-ISAF (N300-N200) carbon blacks. Courtesy of Phillips Petroleum Co.
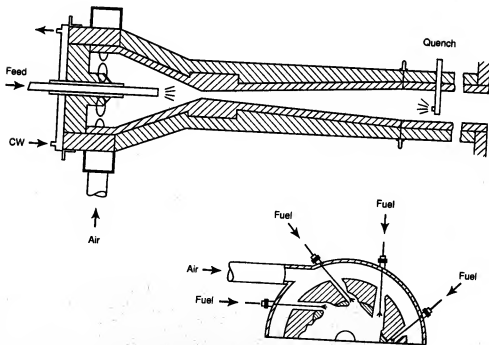

Fig. 10. Columbian reactor for tread blacks. CW = cold water.

quench, heat loss by radiation to atmosphere, and the heat transferred to preheat the primary combustion air. The energy balance for a N300 type of reinforcing grade is shown in Table 4. In this example the thermal efficiency of the process (without tail gas utilization) is 37% for a carbon yield of 0.63 kg/L (5 lb/gal), 61% based on feedstock.

Table 4. Energy Balance for Reinforcing Grade of Carbon Black

Energy input	
energy from feedstock (288°C preheat)	73%
energy from natural gas	23%
air preheat (400°C)	4%
Energy output	
carbon product (heat of combustion)	37%
tail gas (heat of combustion and sensible heat)	39%
heat loss from water quench	14%
heat loss to atmosphere	6%
air preheat (400°C)	4%

THERMAL BLACK PROCESS

Thermal black is a large particle size, low structure carbon black made by the thermal decomposition of natural gas, coke oven gas, or liquid hydrocarbons in the absence of air or flames. Its use in the United States in 1989 was estimated at about 54–68 million kg or about 4% of total consumption. Although at one time, based on cheap natural gas, thermal black was the least expensive of the regular rubber-grade blacks, it is today the most expensive. It is used in rubber and plastics applications for its unique properties of low hardness, high extensibility, low compression set, low hysteresis, and excellent processing. Its main uses are in O-rings and seals, hose, tire innerliners, V-belts, other mechanical goods, and in cross-linked polyethylene for electrical cables.

The thermal black process dates from 1922. The process is cyclic using two refractory-lined cylindrical furnaces or generators about 4 m in diameter and 10 m high. During operation, one generator is being heated with a near stoichiometric ratio of air and off-gas from the make generation whereas the other generator, heated to an average temperature of 1300°C, is fed with natural gas. The cycle between black production and heating is five minutes alternating between generators, resulting in a reasonably continuous flow of product and off-gases to downstream equipment. The effluent gas from the make cycle, which is about 90% hydrogen, carries the black to a quench tower where water sprays lower the temperature before entering the bag filter. The effluent gas is cooled and dehumidified in a water scrubber for use as fuel in the heating cycle. The collected black from the filters is conveyed to a magnetic separator, screened, and hammer-milled. It is then bagged or pelletized. The pelletized form is bagged or sent to bulk

loading facilities. The yield is about 45% of the total carbon content of the process gas with an energy utilization of 2×10^8 J/kg (0.85×10^5 Btu/lb).

ACETYLENE BLACK PROCESS

The high carbon content of acetylene (92%) and its property of decomposing exothermically to carbon and hydrogen make it an attractive raw material for conversion to carbon. Acetylene black is made by a continuous decomposition process at an atmospheric pressure of 800–1000°C in water-cooled metal retorts lined with refractory. The process consists in feeding acetylene into the hot reactors. The exothermic reaction is self-sustaining and requires water cooling to maintain a constant reaction temperature. The carbon black-laden hydrogen stream is then cooled followed by separation of the carbon from the hydrogen tail gas. The tail gas is either flared or used as fuel. After separation from the gas stream acetylene black is very fluffy with a bulk density of only 19 kg/m^3 (1.2 lb/ft^3). It is difficult to compact and resists pelletization. Commercial grades are compressed to various bulk densities up to 200 kg/m^3 (12.5 lbs/ft^3).

A significant use of acetylene black is in dry cell batteries where it contributes low electrical resistance and high capacity. In rubber it gives electrically conductive properties to heater pads, tapes, antistatic belt drives, conveyor belts, and shoe soles. It is also useful in electrically conductive plastics such as electrical magnetic interference (EMI) shielding enclosures. Its contribution to thermal conductivity has been useful in rubber curing bags for tire manufacture. Production capacity for acetylene black in the United States in 1989 was 2.07 million kg from a single plant.

LAMPBLACK PROCESS

The lampblack process has the distinction of being the oldest and most primitive carbon black process still being practiced. The ancient Egyptians and Chinese employed techniques similar to modern methods collecting the lampblack by deposition on cool surfaces. Basically, the process consists of burning various liquid or molten raw materials in large, open, shallow pans 0.5 to 2 m in diameter and 16 cm deep under brick-lined flue enclosures with a restricted air supply. The smoke from the burning pans passes through low velocity settling chambers from which the carbon black is cleared by motor-driven ploughs. In more modern installations the black is separated by cyclones and filters. By varying the size of the burner pans and the amount of combustion air, the particle size and surface area can be controlled within narrow limits. Lampblacks have similar properties to the low area oil-furnace blacks. A typical lampblack has an average particle diameter of 65 nm, a surface area of 22 m^2/g, and a DBPA of 130 mL/100 g. Production is small, mostly in Western and Eastern Europe. Its main use is in

paints, as a tinting pigment where blue tone is desired. In the rubber industry lampblack finds some special applications.

IMPINGEMENT (CHANNEL, ROLLER) PROCESS BLACKS

From World War I to World War II the channel black process made most of the carbon black used worldwide for rubber and pigment applications. The last channel black plant in the United States was closed in 1976. Operations still exist and are even being expanded in Europe. The demise of channel black was caused by environmental problems, cost, smoke pollution, and the rapid development of oil-furnace process grades that were equal or superior to channel black products particularly for use in synthetic rubber tires.

The name channel black came from the steel channel irons used to collect carbon black deposited by small natural gas flames impinging on their surface iron channels. Highly aromatic anthracene oils are used as raw material instead of natural gas. The black is scraped off the rollers, and the off-gases from the steel box enclosed rollers are passed through bag filters where additional black is collected. About half of the black is deposited on the rollers. The purified exhaust gases are vented to the atmosphere. The oils used in this process are high boiling and must be vaporized and conveyed to the large number of small burners by means of a combustible carrier gas. Yield of rubber-grade black is 60% and 10–30% for high quality color grades.

The characteristics of roller process impingement blacks are basically similar to those of channel blacks. They have an acidic pH, a volatile content of about 5%, surface area of about 100 m²/g, and an average particle diameter of 10–30 nm. The smaller particle size grades are used as color (pigment) blacks, and the 30-nm grade is used in rubber.

Characterization and Test Methods

Carbon blacks differ in particle or nodule size, surface area, aggregate size, and aggregate morphology. Surface activity is also a factor in performance, but this feature has been difficult to define or measure. The ultimate dispersible units are aggregates. Aggregate size distribution and morphology determine such properties as surface area, dibutyl phthalate absorption (DBPA), and testing strength. A complete review of the physicochemical characterization of carbon black has been published (21).

Particle Size. The electron microscope is the universally accepted instrument for measuring particle size, aggregate size, and aggregate morphology. Typical electron micrographs of rubber-grade carbon blacks are shown in Figure 11. The grades are classified according to the ASTM D1765 system (2). The first letter N represents a normal rate of cure in rubber, and the first digit represents the average particle size of the carbon black. The last two digits are arbitrarily assigned. Thus N330 is a normal curing grade with a particle diameter range of 26 to 30 nm.

Particle size measurements are made from a negative enlarged to 100,000 diameters (29). Automated image analyzers provide measurements of a variety of
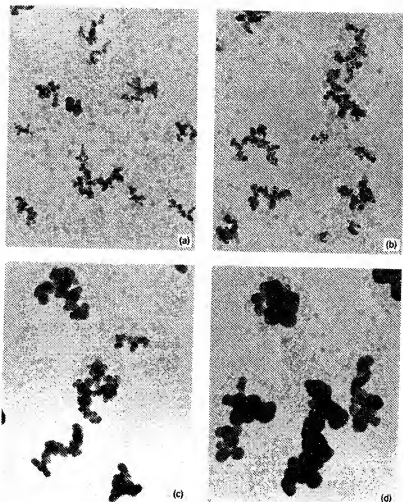
Fig. 11. Electron micrographs of rubber-grade carbon blacks where **(a)** is N110, **(b)** is N220, **(c)** is N550, and **(d)** is N762.

particle and aggregate parameters. Surface areas can be calculated from electron microscope measurements. These are in satisfactory agreement with surface areas determined by nitrogen adsorption measurements. Special pigment blacks and blacks used for electrical conductivity are highly porous, and the surface areas calculated from their particle diameters are very much smaller than those calculated from gas absorption.

Surface Area.   The most important features influencing the performance of carbon blacks are aggregate size and surface area. Surface area is measured by gas- and liquid-phase adsorption techniques, and depends on the amount of adsorbate required to form a surface monolayer. If the area occupied by a single-adsorbate molecule is known, a simple calculation will yield the surface area. A low temperature nitrogen absorption method, based on the original method of

Brunauer, Emmett, and Teller (BET) (30), has been adopted by ASTM as standard method D3037-86 (2).

Liquid-phase adsorption methods are widely used for quality control and specification purposes. The adsorption of iodine from potassium iodide solution is the standard ASTM method D1510-83 (2). The surface area is expressed as the iodine number whose units are milligrams of iodine adsorbed per gram of carbon. It is quite fortuitous that the values of iodine numbers turn out to be about the same as the values for surface areas in square meters per gram by nitrogen adsorption for nonporous carbon blacks.

Another standard industry method for surface area is based on the adsorption of cetyltrimethylammonium bromide (CTAB) from aqueous solution. This is ASTM method D3765-85 (2). This method measures the specific surface area of carbon black exclusive of the internal area contained in micropores that are too small to admit the large CTAB molecules. For rubber-grade nonporous blacks the CTAB method gives excellent agreement with nitrogen surface areas.

Structure and Aggregate Morphology. Structure or aggregate morphology is another important characteristic that influences performance. Structure is determined by aggregate size and shape. These properties affect aggregate packing and the volume of voids in the bulk material. In liquid media structure affects rheological properties such a viscosity and yield point. In rubber, viscosity, extrusion die swell, modulus, and electrical conductivity are affected by structure. For classification and quality control purposes structure is assessed by measurements of void volume, either in the bulk by density or by the absorption of a liquid such as dibutyl phthalate (DBP). The dibutyl phthalate absorption number determination is ASTM method D2414-86 (2). The void volume in the bulk is usually measured under pressure. From the bulk density under a given pressure the volume of voids per unit weight of carbon is calculated.

Tint Strength. Tint strength is another industry method used for the classification of carbon blacks adopted by ASTM as D3265-85 (2). Tint strength is closely related to surface area and decreases with increasing aggregate size. It provides a rough estimate of the reinforcing potential of carbon black in rubber. In this test a small amount of carbon black is mixed with zinc oxide and an oil vehicle to produce a black or gray paste. The reflectance of this paste is measured and compared to the reflectance of a paste made with a reference black. The ratio of the reference black paste reflectance to the sample black multiplied by 100 is the tint strength.

There are many other test methods used to characterize carbon blacks for quality control and specification purposes. Table 5 lists some of these methods which, with a few exceptions, have been adopted by ASTM.

Grades and Applications

U.S. consumption of carbon black in 1988 by various market sectors is shown in Table 6. About 90% of total consumption is in the rubber industry and 69% for tires. About 10% is consumed for other automotive products and 11% for rubber products unrelated to the automotive industry. The automotive industry accounts

Table 5. Special Analytical Test Methods for Carbon Black

Test method	Standard	Comment
iodine adsorption, mg/g	ASTM D1510	amount of iodine adsorbed from aqueous solution as a measure for the specific surface area; not applicable for oxidized or highly porous carbon blacks
N_2 surface area, m^2/g	ASTM D3037	calculated from amount of adsorbed N_2 at liquid nitrogen temperature
CTAB surface area, m^2/g	ASTM D3765	amount of cetytrimethylammonium bromide adsorbed from aqueous solution as measure of specific nonporous (outer) surface area
aggregate dimension	ASTM D3849	determination of aggregate dimensions (unit length, width, etc) by electron microscope image analysis
aggregate size distribution		diameters of equivalent solid spheres that sediment at same rate as aggregates during centrifuging
DBP absorption, mL/100 g	ASTM D2414	determination of the void volume with dibutyl phthalate in a special kneader as measure of structure
void volume, mL/100 g		volume of voids from bulk density measurement under pressure
24M4-DBP absorption, mL/100 g	ASTM D3493	determination of DBP absorption after four repeated compressions at 165 MPa (24,000 psi)
jetness		light absorption of a carbon black paste in linseed oil; determination by visual comparison against standard blacks or by measuring the absolute light emission
tint strength, %	ASTM D3265	ability of a carbon black to darken a white pigment in a linseed oil paste; the tinting strength is the weight percentage of the standard carbon black with respect to the tested black to obtain the same gray tone; different standard white pigments and carbon black concentrations are used according to ASTM
volatiles, %	ASTM D1620	weight loss when calcined at 950°C for 7 min
heating loss (moisture), %	ASTM D1509	weight loss on drying at 125°C for 1 h
pH	ASTM D1512	pH of an aqueous slurry of carbon black; pH is mainly influenced by surface oxides
extractables, %		amount of material which can be extracted by a boiling solvent, usually toluene, in at least 8 h
	ASTM D3392	light absorption–transmission of a 1,2-dichlorobenzene solution of the extracted material

Table 5. (Continued)

Test method	Standard	Comment
ash content, %	ASTM D1506	amount of noncombustible material after burning the carbon black at 675°C
sulfur content, %	ASTM D1619	
sieve residue, %	ASTM D1514	amount of coarse impurities that cannot be purged through a testing sieve by water
bulk density, g/L	ASTM D1513	measure for the densification of carbon black
tamped density, g/L		similar to bulk density; however, void volume is reduced by tamping
pellet size distribution	ASTM D1511	determination by means of sieve shaker
fines content, %	ASTM D1508	only for pelletized blacks; percentage passing through a sieve of 125 μm (mesh) width

for 79% of consumption. Pigment applications account for about 10% of consumption, most of this for plastics and printing inks. Western Europe consumes 74% in tires and other automotive products and almost 20% in other industrial rubber products. Pigment applications in Western Europe and Japan are 5–6% of consumption.

Table 6. U.S. End Use Consumption of Carbon Black[a]

Market sector	Consumption, 10^3 t	Percent of total, %
Rubber		
tires, treads, tubes	927	68.9
other automotive	132	9.8
molded, extruded, industrial products, roofing, etc	148	10.9
Total rubber	*1207*	*89.6*
Nonrubber		
plastics	59	4.4
printing inks	48	3.6
paint	9	0.7
paper	7	0.5
other	16	1.2
Total nonrubber	*139*	*10.4*

[a]1988.

Rubber Goods. A selected list of typical properties, taken from ASTM D1765 of rubber-grade carbon blacks (2), is shown in Table 7. In addition to the assigned ASTM N-numbers, the list includes the old letter designations, pour

densities, structure (DBPA), surface areas, and tint data. The structure/area
relationships of these grades, called the carbon black spectrum, is illustrated in
Figure 12, which shows a diagram of DBPA values versus the nitrogen surface
areas. Closely related grades are easily distinguished. A broad range of structure
is available in the N700-N600 and N300 range of surface areas. Table 8 lists the
principal rubber grades by their N-number classification, general rubber proper-
ties, and typical uses. The behavior of different grades is dominated mainly by
surface area and structure (DBPA). High surface area produces high reinforce-
ment as reflected in high tensile and tear strengths, high resistance to abrasive
wear, higher hysteresis, and poorer dynamic performance. A present day chal-
lenge to carbon black technologists is to optimize the balance between tire wear
and tire hysteresis or the rolling resistance. Some progress on this problem has
been made by using new furnace designs and other process variables that broaden
the aggregate size distributions and lower the tint strength while maintaining
surface area, structure, and reinforcement (31,32).

Fig. 12.   Rubber grades carbon black spectrum.
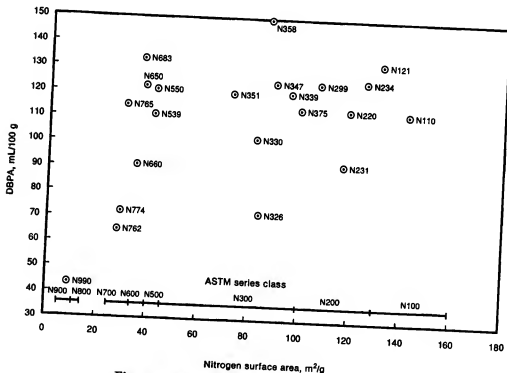
The consumption of the various carbon black grades can be divided into
tread grades for tire reinforcement and nontread grades for nontread tire use and
other rubber applications. Table 9 shows the distribution of production of types
for these uses. In the United States 55% production is for tread grades. In Western
Europe tread-grade production is 64%, and in Japan it is 60%.

Table 7. Typical Properties Rubber-Grade Carbon Blacks[a]

ASTM designation	Former industry designation[b]	I2 absorption number (D1510), g/kg	DBPA[c] (D2414), mL/100 g	DBPA[c] (compressed sample) (D3493), mL/100 g	CTAB[d] surface area (D3765), m²/g	Nitrogen surface area (D3037), m²/g	Tinting strength (D3265)	Pour density (D1513), kg/m³
N110	SAF	145	112	98	126	143	124	335
N121	SAF-HS	121	132	112	121	132	121	320
N220	ISAF	121	114	100	111	119	115	345
N231	ISAF-LM	121	92	86	108	117	117	390
N234	ISAF-HS	120	125	100	119	126	124	320
N299	ISAF-HS	108	124	105	104	108	113	335
N326	HAF-LS	82	72	69	83	84	112	465
N330	HAF	82	102	88	83	83	103	375
N339	HAF-HS	90	120	101	96	90	110	345
N347	HAF-HS	90	124	100	88	88	103	335
N351	HAF-HS	68	120	97	74	73	100	345
N358	HAF-HS	84	150	112	98	87	99	290
N375	HAF-HS	90	114	97	98	100	115	345
N539	FEF	43	111	84	41	41	0	385
N550	FEF	43	121	88	42	42	0	360
N650	GPF-HS	36	122	87	38	38	0	370
N660	GPF	36	90	75	35	35	0	425
N683	GPF-HS	35	133	0	39	37	0	335
N762	SRF	27	65	57	29	28	0	505
N765	SRF-HS	31	115	86	33	31	0	375
N774	SRF	29	72	62	29	29	0	495
N990	MT	0	43	40	9	9	0	0

[a] ASTM D1765.
[b] SAF = super abrasion furnace; ISAF = intermediate super abrasion furnace; HAF = high abrasion furnace; FEF = fast extrusion furnace; GPF = general purpose furnace; SRF = semireinforcing furnace; MT = medium thermal; HS = high structure; LS = low structure; LM = low modulus.
[c] Dibutyl phthalate [84-74-2] absorption.
[d] Cetyl trimethylammonium bromide [57-09-0].

Table 8. Applications of Principal Rubber-Grade Carbon Blacks

Designation	General rubber properties	Typical uses
N110, N121	high abrasion resistance	special tire treads, airplane, off-the-road racing
N220, N299, N234	high abrasion resistance, good processing	passenger, off-the-road, special service tire treads
N339, N347, N375, N330	high abrasion resistance, easy processing, good abrasion resistance	standard tire treads, rail pads, solid wheels, mats, tire belt, sidewall, carcass retread compounds
N326	low modulus, good tear strength, good fatigue, good flex cracking resistance	tire belt, carcass, sidewall compounds, bushings, weather strips, hoses
N550	high modulus, high hardness, low die swell, smooth extrusion	tire innerliners, carcass, sidewall, innertubes, hose, extruded goods, v-belts
N650	high modulus, high hardness, low die swell, smooth extrusion	tire innerliners, carcass, belt, sidewall compounds, seals, friction compounds, sheeting
N660	high modulus, high hardness, smooth extrusion	carcass, sidewall, bead compounds, innerliners, seals, cable jackets, hose, soling, EPDM compounds
N762	high elongation and resilience, low compression set	mechanical goods, footwear, innertubes, innerliners, mats

Table 9. Carbon Black Production[a] by Grade, 10^3 t

	United States	Western Europe	Japan
N100	35	28	37.1
N200	158	161	118
N300	555	528	300
Total tread grades	*748*	*717*	*418*
percent	55.2	63.8	59.5
N500	120	153	136
N600	326	137	87
N700	129	103	29
N900 (thermal)	23		9
Total nontread grades	*598*	*393*	*261*
percent	44.1	35.0	37.1
other grades			
acetylene	9.1	14	24
Total carbon black	*1355*	*1124*	*703*

[a] 1988.

1062

Special-Grade Carbon Blacks. In 1988 over 10% of U.S. consumption of carbon black was for nonrubber applications, ie, special blacks. In Europe and Japan about 5% is consumed for these uses. Most of the special black grades are manufactured by methods to meet specific product specifications required for their end uses. They sell for a higher average price than the rubber grades. These markets have been growing at an average annual rate twice that of the rubber black grades. Of increasing importance in recent years have been applications in plastics to improve weathering resistance and to impart antistatic and electrically conductive properties.

About 42% of special blacks are used in plastics, 35% in printing inks, 7% in paper, and 16% in miscellaneous applications. News inks account for most of the printing ink market. Electrical applications have been taking an increasing share of the plastics market. Medium and high color grades, in their normal and surface-oxidized versions, are used in enamels, lacquers, and plastics for their extreme jetness. Typical properties of special grades of furnace blacks are listed in Table 10. The list is divided into normal furnace grades and surface oxidized grades. Increased surface oxidation decreases viscosity, improves dispersion, and increases the flow behavior in many liquid systems. The volatile content is an indication of the degree of surface oxidation. To improve dispersion and flow, special blacks generally are produced at lower structural (DBPA) and bulk density values than rubber-grade carbon blacks.

Table 11 lists the types and applications of special pigment-grade carbon blacks. Included in this list are thermal black and lampblack. Over 40 special black grades have been developed based on the furnace process having a broad range of surface areas, from 20 m^2/g to over 1500 m^2/g. The lower surface area products are used in printing inks and tinting. The high area, more expensive products find use in high color enamels and lacquers.

Electrically Conductive Grades. An important application of carbon black is to produce electrically conductive and antistatic polymer composites. These applications include antistatic carpet backing and floor tile, electrical heating elements, high voltage cable semiconductive shields, video tapes and disks, and EMI shielding. The electrical conductivity of bulk carbon black under compression is in the range of 0.02 to 0.5 ohm·cm. The conductivity of conductive carbon black-filled rubber and plastics is in the range of 1 to 10^8 ohm·cm. There is no clear relationship between bulk conductivity and compound conductivity. The main variable determining compound conductivity is the carbon black concentration. At high enough concentrations all carbon blacks can produce compound resistivities of about 1.0 ohm·cm. For superconductive carbon black this concentration is 7–8% and for thermal black the required concentration is 65–70%. Figure 13 shows the concentration/resistivity relationships of selected carbon blacks covering the complete range of rubber and conductive grades (33). It can be seen that there is a critical concentration for each grade of carbon black above which the resistivity drops precipitously. This is often referred to as the percolation concentration.

The main carbon black characteristics determining its conductive behavior are surface area, aggregate morphology, and degree of graphitization or crystallinity. The high conductivity of acetylene black is attributed to its highly developed structure and its crystallinity. High conductivity furnace-grade blacks have

Table 10. Furnace Process Special Grades for Pigment Applications in Inks, Plastics, Paints, and Paper

Industry classification	N_2 surface area, m²/g	Particle diameter, nm	DBPA[a], mL/100 g		Bulk density, g/L		Nigrometer[b] index	Tinting strength	Volatile, %	pH
			Fluffy	Pellets	Fluffy	Pellets				
high color	250–300	14–15	70–75	60–65	50–300	400–550	65–76	117–124	1.2–2.0	7–10
medium color	150–220	16–24	47–122	46–117	130–300	390–550	74–78	118–124	1.0–1.5	8–10
regular color	45–140	20–37	42–125	42–124	176–420	350–600	84–93	73–119	0.9–1.5	6–10
low color	25–45	41–75	71	64–120	256	352–512	94–99	48–69	0.6–0.9	8–10
Surface oxidized grades										
high color	400–600	10–20	121	105			64	100–135	8.0–9.5	2.0–3.3
medium color (long flow)	100–38	23–24	49–60	55	240–360	530	83–84	112–135	3.5–5.0	2.5–4.0
medium color (medium flow)	96–110	25	49–72	70	225–360	480	84	112–114	2.5–3.5	4.0–4.5
low color	30–40	50–56	48–93		260–500		92–100	64	3.5	3.0

[a]Dibutyl phthalate absorption.
[b]A method for measuring the diffuse reflectance from a black paste with a black tile standard. The low numbers represent the jettest or most intense black grades.

Table 11. Types and Applications of Special Pigment Grades of Carbon Blacks

| Type	Surface area, m^2/g	DBPA[a], mL/100 g	Volatile content, %	Uses
Normal grades				
high color	230–560	50–120	2	high jetness for alkyl and acrylic enamels, lacquers, and plastics
medium color	220–220	70–120	1–1.5	medium jetness and good dispersion for paints and plastics; ultraviolet and weathering protection for plastics
regular color	80–140	60–114	1–1.5	for general pigment applications in inks, paints, plastics, and paper; gives ultraviolet protection in plastics, high tint, jetness, gloss, and dispersibility in inks and paints
	46	60	1.0	good tinting strength, blue tone, low viscosity; used in gravure and carbon paper inks, paints, and plastics
	45–85	73–100	1.0	main use is in inks; standard and offset news inks
low color	25–42	64–120	1.0	excellent tinting black-blue tone; used for inks-gravure, one-time carbon paper inks; also for paints, sealants, plastics, and cements
thermal blacks	7–15	30–35	<0.5	tinting-blue tone; plastics and utility paints
lamp blacks	20–95	100–160	0.4–0.9	paints for tinting-blue tone
Surface oxidized grades				
high color	400–600	105–121	8.0–9.5	used for maximum jetness in lacquers, coatings, plastics, fibers, record disks
medium color, long flow	138	55–60	5	used in lithographic, letterpress, carbon paper, and typewriter ribbon inks; high jetness, excellent flow, low viscosity, high tinting strength, gloss, and good dispersability
medium color, long flow	96	70	2.5	used for gloss printing and carbon paper inks; excellent jetness, dispersibility; tinting strength, and gloss in paints
low color	30–40	48–93	3.5	used for tinting where flooding is a problem; easy dispersion

[a]Dibutyl phthalate absorption.
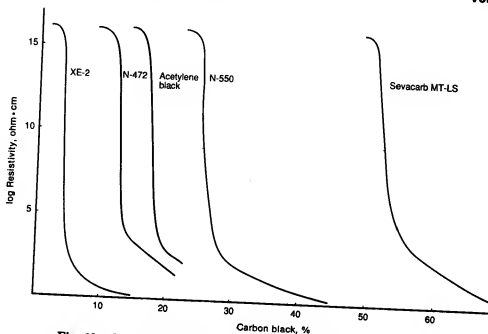
Fig. 13.   Electrical resistivity versus carbon black concentration.

very high surface areas and structures (DBPA). Superconductive blacks, by-products of synthesis gas manufacture, have very high surface areas, highly developed structures, and a high degree of crystallinity.

The effect of surface area on conductivity was shown for channel-grade blacks in 1949 (34) and the surface area relationship to conductivity for furnace blacks in 1954 (35). High surface area is associated with increasing surface roughness and internal porosity rather than decreased particle or nodule size. Because of the decreased density of the aggregates resulting from the porosity of high surface area conductive blacks, they possess a larger number of aggregates per unit weight. At a given weight concentration, closer packing of aggregates increases conductivity. Crystallinity increases with high porosity contributing to high conductivity. The crystallinity increase results from the burnout of the more amorphous regions of the aggregate during manufacture.

The mechanism of electrical conduction in composites occurs by a process of electron tunneling through the polymer phase (36). Electrons tunnel from the black aggregates to their nearest neighbor. The resistivity of vulcanizates is a function of the average distance between aggregates (37). In addition to carbon black concentration, this gap distance depends on particle size, surface area, and aggregate morphology.

There are a number of publications on the properties and applications of electrically conductive carbon blacks (38–40). Figure 14 shows the electron micrographs of two grades of electrically conductive carbon blacks. The furnace blacks have the particle size of N200-N300 types. Their high surface areas indicate high internal porosity. Table 12 shows typical data and uses for eight electrically

conductive grades of carbon black and by-product carbons. The large differences between the nitrogen surface areas and the areas measured by cetyltrimethyl-ammonium bromide (CTAB) absorption are because of internal porosity. The CTAB molecules are so large that they do not penetrate the pores available to the nitrogen molecules. The large, bulky aggregates, the high porosities, and low aggregate densities of the electrical grades produce high DBPA values, much larger than for normal furnace blacks.
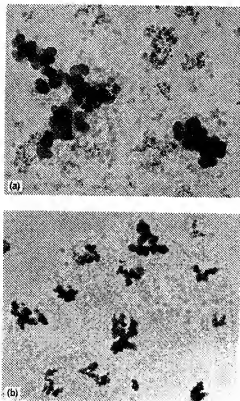

Fig. 14. Electron micrographs of electrically conductive grades of carbon black where (a) is Vulcan XC-72 (Cabot) (100,000 ×) and (b) is Vulcan P (Cabot) (100,000 ×).

Carbon Black Manufacture and Market

Manufacturers and Productions. The consumption of carbon black in the United States reached a peak of 1,506,000 t during the beginning of the oil crisis in 1973. Then consumption decreased to 1,210,000 t in 1989. A number of events have contributed to decreased consumption by the rubber and tire industries including tire radialization, increased tire mileage, downsizing of tires, and increased imports of foreign cars. The negative influence of these events have pretty much run their course, and during the last 10 years there has been a modest growth in

Table 12. Typical Data and Uses for Electrically Conductive Grades of Carbon Blacks

Type	Particle diameter, nm	N_2 Surface area, m^2/g	CTAB Surface area, m^2/g	DBPA[a] mL/100 g	Tinting strength, D3265	Uses
acetylene black	42	64		300	52	high voltage semiconductive shields, conductive rubber, and plastics
conductive furnace (CFJN293)	22	145	114	100	117	conductive rubber and plastics, carpet backing
conductive furnace (CFJN742)	22	270	145	178	82	conductive and antistatic rubber and plastics products
superconductive furnace (SCF)		1475 1000		330 245	163	electromagnetic interference shielding (EMI) compounds, videodisks, tapes, etc
synthesis gas by-product carbon	30	800 1000 1250	620	365 400 495	124	EMI, videodisks, PTC[b] compounds (for heating tapes)

[a]Dibutyl phthalate absorption.
[b]PTC = positive temperature coefficient.

carbon black production. Production for the period 1973–1989 is shown in Figure 15.
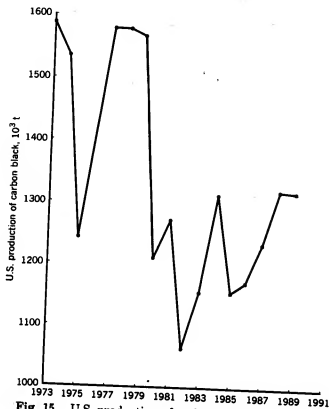

Fig. 15.   U.S. production of carbon black (1973–1989).

The shrinkage in demand has resulted in a restructuring of the carbon black industry. Several of the principal multinational oil companies have left the business including Ashland, Cities Service Co., Phillips, and Conoco. Some plants have changed ownership. In the United States this has increased the production capacities of Degussa, Sid Richardson, and Huber. Today's U.S. industry consists of six principal producers. Rated capacities of the six U.S. manufacturers is shown in Table 13. Cabot Corp. and Columbian Chemicals are the leading producers, followed by Degussa, Sid Richardson, J. M. Huber Corp., and Witco. A survey of the future markets and present structure of the carbon black industry has been presented (1).

World carbon black rated capacities are shown in Table 14. North America has the largest capacity. Europe, Southeast Asia, and Russia/Eastern Europe have about equal capacities and Africa and the Middle East have only small production. The growth areas are predicted to be Southeast Asia and the Russia/Eastern Europe markets. The capacities for certain areas such as China and Russia/Eastern Europe should be taken as rough estimates.

Table 13. U.S. Carbon Black Manufacturers Nameplate Capacities, 1988

Manufacturers	Capacity 10^3 t	U.S. capacity, %
Cabot Corp.		
Franklin, La.	153	
Pampa, Tex.	32	
Parkersburg, W.Va.	83	
Ville Platte, La.	126	
Total	*394*	25
Columbian Chemicals		
El Dorado, Ark.	75	
North Bend, La.	121	
Hickok, Kans.	38	
Moundsville, W.Va.	82	
Total	*316*	20
Degussa		
Arkansas Pass, Tex.	82	
Ivanhoe, La.	100	
Belpre, Ohio	55	
Total	*237*	15
Sid Richardson Co.		
Addis, La.	62	
Big Spring, Tex.	57	
Borger, Tex.	125	
Total	*244*	15.5
J. M. Huber Corp.		
Orange, Tex.	62	
Baytown, Tex.	102	
Borger, Tex.—Furnace	57	
Thermal	23	
Total	*244*	15.5
Witco Corp.		
Phenix City, Ala.	25	
Ponca City, Okla.	68	
Sunray, Tex.	46	
Total	*139*	8.8
Total U.S. capacity	*1574*	

Product Safety

The safety aspects of carbon black have been the subject of a number of reviews and articles (41–43). The manufacture of carbon results in trace amounts of organic and inorganic impurities. These impurities have been suspected of causing potential health problems. Of particular concern have been the salts of toxic metals and adsorbed polynuclear aromatic hydrocarbons (PNAs). A few of the polyaromatic hydrocarbons are known to be mutagens and/or animal carcinogens. The solvent extract of furnace blacks is in the range of 300 to 2000 ppm (0.03–0.20%). Most of this extract consists of 10–15 organic compounds, the

Table 14. World Carbon Black Capacities by Region and Country, 1988

Region	Number of plants	Estimated capacity, 10^3 t
North America		
United States	22[a]	1565[a]
Canada	3[a]	182
Mexico	2	158
Total	*27*	*1095*
South America		
Argentina	1	48
Brazil	3	199
Colombia	2	36
Peru	1	8
Venezuela	1	40
Total	*8*	*331*
Europe		
Great Britain	2	140
France	3	239
Germany	5	385
The Netherlands	2	120
Italy	3	191
Portugal	1	21
Spain	3	104
Sweden	1	33
Total	*20*	*1233*
Australia/South East Asia		
Australia	2	76
India	7	153
China[b]	30–45	500
Japan	12	766
South Korea	3	170
Malaysia	1	23
Pakistan	1	10
Philippines	1	15
Taiwan	1	53
Thailand	1	20
Total	*32*	*1786*
Africa	2	75
Middle East		
Iran	1	15
Turkey	1	35
Total	*2*	*50*
Russia/Eastern Europe		
Russia[b]	14	1300
Yugoslavia	1	36
Poland		58
Romania		130
Czechoslovakia		64
Total		*1588*
Total world capacity		*6968*

[a]Includes one thermal black plant (capacity, 25,000 t).
[b]Estimate.

majority of which are not genotoxic. One compound that is toxic is benzo [α]
pyrene [50-32-8], BαP, often used as an indicator of potential hazard. BαP ranges
from 0 to 50 ppm and is less than one percent of the total extract. There have been
a number of studies initiated by the U.S. carbon black industry to examine the
health effects of various commercial carbon blacks and their benzene extracts.
Tests have been made using laboratory animals. Investigations on absorption and
elution effects in stomach fluids and human blood have been conducted. Although
the solvent extracts of carbon black do show toxic properties, the aqueous
systems of concern in humans show no elution of BαP and no toxic properties. The
BαP is believed to be so strongly absorbed on the surface of carbon black and in
such high dilution that it is inactive in animal testing for carcinogenicity. Statis-
tical studies on the frequency of cancer of long-term employees in a carbon black
plant covering a period of 17 years (1939–1956) has been reported (44). There is no
evidence of increased cancer risk from exposure to industrial carbon blacks. The
scientific literature based on animal research as well as observations on carbon
black plant employees show no evidence of detrimental health effects. Most
studies of carbon black dust inhalation and intratracheal administration with
animals indicate that carbon black is not carcinogenic. OSHA regulations for
carbon black dust concentrations call for an average exposure level over a given
time period of not more than 3.5 mg/m^3.

Environmental Aspects

The carbon black industry takes extreme efforts to confine product during all
stages of manufacture (45). Highly efficient bag filters are used to collect the
product. After collection the fluffy carbon black is densified and pelletized to
minimize dusting problems during shipping and use.
    The process gases from the filters consist of nitrogen, carbon monoxide,
carbon dioxide, hydrogen, water, small amounts of hydrogen sulfide, and other
sulfur- and nitrogen-containing gases. In the past the process gases have been
flared. Process gas is used as a fuel for in-plant heat needs, and where local
conditions warrant, it may be burned to generate steam or power.

BIBLIOGRAPHY

"Carbon Black" under "Carbon" in *ECT* 1st ed., Vol. 3, pp. 34–65, and Suppl. 1, pp. 130–144,
by W. R. Smith, Godfrey L. Cabot, Inc.; "Acetylene Black" in *ECT* 1st ed., Vol. 3, pp. 66–69,
by B. P. Buckley, Shawinigan Chemicals Ltd.; "Carbon Black" under "Carbon" in *ECT*
2nd ed., Vol. 4, pp. 243–282, by W. R. Smith, Cabot Corp., D. C. Bean, "Acetylene Black",
Shawinigan Chemicals Ltd.; "Carbon Black" under "Carbon" in *ECT* 3rd ed., Vol. 4, pp.
631–666, by E. M. Dannenberg, Cabot Corp.

 1. A. McNeish, *Future Markets for Carbon Black*, Paper No. 48, Rubber Division, ACS
    Meeting, Detroit, Mich., 1989.
 2. *ASTM Standards for Carbon Black*, American Society of Testing Materials, Philadel-
    phia, Pa., 1987.
 3. J. B. Donnet and A. Voet, *Carbon Black*, Marcel Dekker, Inc., New York, 1976, pp.
    1–19.

4. E. M. Dannenberg, "The Carbon Black Industry: Over a Century of Progress," *Rubber World Mag. Spec. Pub.—Rubber Div. 75th Anniv.* (1907–1984).

5. C. R. Houska and B. E. Warren, *J. Appl. Physics* **25**, 1503 (1954).

6. B. E. Warren, *Phys. Rev.* **59**, 693 (1941).

7. L. L. Ban and W. M. Hess, *Abstr. 9th Bienn. Conf. Carbon*, 162 (1969).

8. A. I. Medalia, in E. K. Sichel, ed., *Carbon Black—Polymer Composites*, Marcel Dekker, Inc., New York, 1982, Chapt. 1.

9. E. Redman, F. A. Heckman, and J. E. Connelly, Paper No. 14, Rubber Division, ACS Meeting, Chicago, Ill., 1977.

10. F. Lyon and K. Burgess, in J. I. Kroschwitz, ed., *Encyclopedia of Polymer Science and Engineering*, John Wiley & Sons, Inc., New York, 1985, pp. 623–640.

11. U.S. Pat. 4,071,496 (Jan. 31, 1978), G. Kraus and H. R. Hunt (to Phillips Petroleum Co.).

12. U.S. Pat. 4,478,973 (Oct. 23, 1984), S. Misono and H. Suzuki (to Tokai Carbon Co., Ltd.).

13. A. Patel and K. Lee, *Elastomerics*, **22** (Apr. 1990).

14. C. W. Sweitzer and W. C. Goodrich, *Rubber Age* **55**, 169 (1944).

15. *ASTM Standards for Carbon Black*, D2414-86, American Society for Testing and Materials, Philadelphia, Pa., 1987.

16. *ASTM Standards for Carbon Black*, D3493-86, American Society for Testing and Materials, Philadelphia, Pa., 1987.

17. W. M. Hess, L. L. Bann, and G. C. MacDonald, *Rubber Chem. Technol.* **42**, 1209 (1969).

18. F. A. Heckman and A. Medalia, *J. Inst. Rubber Ind.* **3**, 66 (1969).

19. B. B. Boonstra, H. Cochrane, and E. M. Dannenberg, *Rubber Chem. Technol.* **48**, 4 (1975).

20. A. M. Gessler, W. M. Hess, and A. Medalia, *Plast. Rubber Process.* **3** (1978).

21. J. Janzen, *Rubber Chem. Technol.* **56**(3), 669 (1982).

22. U.S. Pat. 3,010,794 (Nov. 28, 1961), G. F. Frianf and B. Thorley (to Cabot Corp.).

23. M. Faraday, in W. Crooker, ed., *The Chemical History of a Candle*, Viking Press, New York, 1960.

24. *Chem. Eng. News*, 30 (Feb. 5, 1990).

25. E. M. Dannenberg, unpublished data, 1991.

26. H. P. Palmer and C. F. Cullis, in P. L. Walker, ed., *Chemistry and Physics of Carbon*, Vol. 1, Marcel Dekker, Inc., New York, 1965, p. 265.

27. J. LaHaye and G. Prado, in P. L. Walker and P. A. Thrower, eds., *Chemistry and Physics of Carbon*, Vol. 14, Marcel Dekker, Inc., New York, 1965, p. 167.

28. D. Rivin, *Rubber Chem. Technol.* **56**(3), 709 (1982).

29. B. Schubert, F. Lyon, and F. Ford, *Encyclopedia of Industrial Chemical Analysis*, Vol. 8, John Wiley & Sons, Inc., New York, 1969, p. 180.

30. S. Brunauer, P. H. E. Emmett, and J. Teller, *J. Am. Chem. Soc.* **60**, 310 (1938).

31. U.S. Pats. 4,071,496 (Jan. 31, 1978); 4,241,022 (Dec. 23, 1980); 4,267,160 (Feb. 16, 1982), G. Kraus and H. R. Hunt (to Phillips Petroleum Co.).

32. U.S. Pat. 4,478,973 (Oct. 23, 1984), S. Misono and H. Suzuki (to Tokai Carbon Co., Ltd.).

33. N. Prost, *Conductive Rubber Compounds—Compounding with Extra-Conductive Carbon Black*, Phillips Petroleum Chemicals Bulletin, June 1985.

34. E. M. Dannenberg and H. J. Collyer, *Ind. Eng. Chem.* **44**, 1607 (1949).

35. B. B. Boonstra and E. M. Dannenberg, *Ind. Eng. Chem.* **46**, 218 (1954).

36. L. R. VanBeck and B. L. Van Puh, *J. Appl. Polym. Sci.* **6**, 651 (1962).

37. M. H. Polley and B. B. Boonstra, *Rubber Chem. Technol.* **3**, 170 (1959).

38. R. R. Juengel, *Carbon Black Selection for Conductive Rubber Compounds*, Paper No. 78, Rubber Division, ACS Meeting, Los Angeles, Calif., Apr. 1985.

39. W. F. Verkelst, *Electroconductive Black—Possibilities and Limitations*, Scandanavian Rubber Conference, Akzo Chimie, Copenhagen, Denmark, 1985.

40. N. Probst and J. C. Bouquin, *Extra Conductive Black Printex XE2 and Its Multiple*

Facets in The Polymer Industry, Paper No. 59, Rubber Division, ALS Meeting, Montreal, Canada, May 1987.
41. D. Rivin and R. G. Smith, *Rubber Chem. Technol.* **55**, 3 (1982).
42. D. Rivin, *Dangerous Prop. Ind. Mater. Rep.* **5**, 1 (1985).
43. D. Rivin, *The Handbook of Environmental Chemistry,* Vol. 3, Part D, Springer-Verlag, Berlin, Germany, 1986, pp. 101–158.
44. T. H. Ingalls, *Arch. Environ. Health* **2**, 429 (1961).
45. E. M. Dannenberg, *Rubber Age* **108**, 4 (1976).

ELI M. DANNENBERG
Consultant

LYN PAQUIN
HARRY GWINNELL
Cabot Corporation

EVIDENCE APPENDIX II

Encyclopedia of

Industrial Chemical

Analysis

Editor-in-Chief: FOSTER DEE SNELL
Executive Editor: LESLIE S. ETTRE

Volume 8
Bromine
to
Carrier Gas and
Vacuum Fusion Methods

ENCYCLOPEDIA

OF INDUSTRIAL

CHEMICAL

ANALYSIS

Edited by Foster Dee Snell *and* Leslie S Ettre

VOLUME 8

Bromine

to

Carrier Gas and Vacuum

Fusion Methods

anion analysis is also necessary. The anions present in carbon black ash are most typically sulfates and oxides. The relative levels of each are dependent on the amount of sulfur present in the carbon black.

Very low ash (as well as low residue) is particularly important for conductive carbon blacks which are used in plastics for electrical cables. Most critical is the semiconductor strand shield compound that is employed to reduce electrical stress as a thin layer between the metal conductor and insulator. The presence of metallic ions or particulate impurities at the strand shield-insulator interface can promote premature cable failure.

References

1. Donnet, J. B. and Voet, A., *"Carbon Black, Physics, Chemistry, and Elastomer Reinforcement"*, Marcel Dekker, Inc., New York and Basel, 1976.
2. Dannenberg, E. M., Carbon Blacks, in *"Kirk-Othmer Encyclopedia"*, Vol.4, John Wiley and Sons, New York, p. 631, 1978.
3. Medalia, A. I, and Rivin, D., *"Carbon Black, Characterization of Powder Surfaces"*, G. D. Parfitt and K. S. W. Sing eds., Academic Press, New York, New York, p. 279, 1976.
4. Kraus, G., ed., *"Reinforcement of Elastomers"*, Interscience, New York, 1965.
5. Gessler, A. M., Hess, W. M. and Medalia, A. I., *Plastics and Rubber Processing*, 3, 1 (1978).
6. Heckman, F. A., *Rubber Chem. Technol.*, 37, 1245 (1964).
7. Ban, L. L. and Hess, W. M., *"Interactions Entre Les Elastomeres et les Surfaces Solides Ayant Une Action Renforcante"*, Colloques Internationaux Du CNRS, France, September 24-26, p. 81, 1973.
8. Studebaker, M. L., *Rubber Chem. Technol.*, 30, 1400 (1957).
9. Rivin, D., *Rubber Chem. Technol.*, 44, 307 (1971).
X 10. Schubert, B., Ford, F. P. and Lyon, F., *"Encyclopedia of Industrial Chemical Analysis"*, Vol.8, John Wiley and Sons, Inc., p.179, 1969.
11. Jansen, J., *Rubber Chem. Technol.*, 55, 669 (1982).
12. Wiegand, W. B., *Canadian Chemical Journal*, 4, 160 (1920).
13. Green, H., *Chem. and Met. Eng.*, 28, 53 (1923).
14. Spear, E. B., *Colloid Symposium Monographs*, 1, 321 (1923).
15. Goodwin, N. and Park, C. R., *Ind. Eng. Chem.*, 20, 621 (1928).
16. Grenquist, E. A., *Ind. Eng. Chem.*, 21, 665 (1929).
17. Parkinson, D. N., *Trans. Inst. Rubber Ind.*, 5, 263 (1930).
18. Knoll, M. and Ruska, E., *Ann. der Physik*, 12, 607 (1932).
19. Knoll, M. and Ruska, E., *Z. Physik*, 78, 318 (1932).
20. Prebus, A. F. and Hillier, J., *Can. J. Research*, 17, 49 (1939).
21. *Columbian Colloidal Carbons* Vol. II; *Rubber Chem. Technol*, 14, 52 (1940).

masstone. The effect of structure on the masstone–particle size correlation is also apparent. Structure has a depressing effect on blackness, with maximum masstone occurring at minimum structure.

Tinting Strength. Another optical test in common usage to estimate carbon black particle diameter is tinting strength which measures the ability of a carbon black to blacken a standard white pigment–oil mixture. A typical tinctorial paste used in the carbon black industry for particle size control consists of 20 mg of carbon and 2.0 g of zinc oxide mulled in 2.7 ml of mineral oil. A quantitative comparison of carbon blacks can be made by determining the amount of a standard black that is needed to match the strength of the test sample.

Tinting strength is far more limited in scope than masstone in predicting the particle diameter of carbon black. Tinting strength increases with decreasing particle size. Below 150 Å, there is little change in tint with decreasing particle size. This limitation on tinting strength is believed due to the increasing magnitude of the difference in particle sizes between the white pigment and the carbon blacks. The very small particle carbons are effectively hidden by the large white pigment particles.

TOTAL SURFACE AREA

Nitrogen Adsorption. Carbon blacks differ not only in regard to their external surface areas but also in their internal surface areas. In general, rubber grade carbons have smooth, impervious surfaces. With such carbons, if structure is at a minimum, the surface areas determined by measuring particle diameters from electron microscope pictures and gas adsorption measurements should be essentially equal. The presence of structure, however, or capillary connections to the interior of the particles leads to error in the calculation of surface area from particle diameter, and gas adsorption techniques are required for valid total surface area measurements.
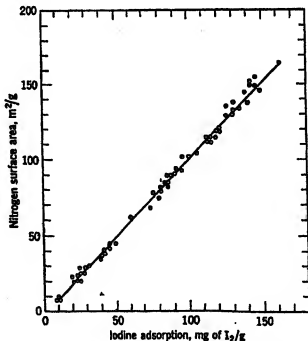

Fig. 7. Nitrogen-iodine surface correlation for furnace blacks.

The total surface area of carbon blacks is generally determined by gas adsorption techniques based on the Brunauer, Emmett, and Teller (BET) procedure (8). This method, based on low-temperature nitrogen adsorption, has been critically reviewed and criticized on theoretical grounds (9), but it has proved to be a very reliable one and has gained general acceptance.

A measure of the internal surface area, or roughness factor, of carbon black can be obtained by taking the ratio of the surface area measured by nitrogen adsorption to the area computed from electron micrographs (10).

Iodine Adsorption. There have been many control tests for surface area developed and satisfactorily correlated with surface areas calculated from gas adsorption, but because of its simplicity and precision, iodine adsorption (11) has gained wide acceptance as a surface area control test. This is true in the rubber industry where the low-volatile, nonporous carbons are particularly well suited for iodine control. The iodine adsorption–nitrogen (BET) surface area correlation for rubber-grade carbons is shown in Figure 7.

Deviations from linearity in the BET nitrogen–iodine adsorption correlation occur with carbons having a highly unsaturated oil on the surface or an oxygen volatile content per unit surface area significantly greater than the rubber grade furnace carbons. Unsaturated oils are present in significant amounts only on thermal carbons, but the high oxygen content of channel carbons and oxidized furnace carbons excludes all these carbons from satisfactory surface area estimates by the use of iodine adsorption.

The primary factor responsible for the poor correlation with oxidized carbons is the highly polar channel black surface which preferentially adsorbs water from the aqueous potassium iodide–iodine system, resulting in low iodine adsorption; the higher the oxygen content, the greater the deviation from linearity for the nitrogen–iodine adsorption correlation. A secondary, but minor factor, responsible for the relatively low iodine adsorption values for oxidized carbon black is the oxidation of iodide to iodine by the oxygenated carbon surface.

<center>STRUCTURE</center>

Technologists have come to use the term *structure* in more than one sense when discussing carbon black. The internal crystalline nature of carbon black particles has been referred to, in general, as the *microstructure*. More frequently, structure refers to the tendency of carbon black particles to form in fibrous or clustered aggregates. This property of carbon black has also been termed *reticulate structure*.

Microstructure. The early x-ray diffraction work of Biscoe and Warren (12) suggested that carbon black particles are composed of small randomly oriented graphite crystallites in which the layer planes are stacked roughly parallel and equidistant but in rotational disorder about the c axis (turbostratic orientation). They defined carbon black as an intermediate form of matter, neither completely crystalline nor completely amorphous.

Later, Hall, using dark field electron microscopy, suggested that crystallite orientation might not be completely random in carbon black (13). His studies on a thermal black indicated that the crystallites were oriented—the graphite layers being roughly parallel to the surface of the particles. This finding was subsequently supported by the studies of Donnet and Bouland (14) and Heckman (15) by means of electron microscope studies on severely oxidized carbon blacks. Heckman and Harling (16)

54. P. H. Emmett and S. Brunauer, *J. Am. Chem. Soc.* 59, 1553–1564 (1937).
55. W. Barr and V. Anhorn, *Scientific and Industrial Glass Blowing and Laboratory Techniques*, Instruments Publishing Co., Pittsburgh, Pa., 1949, Chap. XII.
56. P. A. Faeth and C. B. Willingham, *Assembly, Calibration, and Operation of a Gas Adsorption Apparatus for the Measurement of Surface Area, Pore Volume Distribution, and Density of Finely Divided Solids*, Mellon Institute of Industrial Research, Pittsburgh, Pa., Sept. 1955.
57. W. V. Loebenstein and V. R. Deitz, *J. Res. Natl. Bur. Std.* 46, 51–55 (1951).
58. J. Kremens, J. S. Lagarius, and V. R. Deitz, "A New Approach to Surface Area Determinations by Selective Gas Adsorption in a Nitrogen–Helium System," *Paper No. 110, Pittsburgh Conference on Analytical Chemistry and Applied Spectroscopy, March 1–5, 1965*, Reprint No. 232, American Instrument Co., Inc., Silver Spring, Md.
59. *Aminco-Deitz Sor-BET Cat. No. 5–7300 and 5–7302 Instruction No. 839*, American Instrument Co., Inc., Silver Spring, Md., Nov. 1965.
60. F. M. Nelsen and F. T. Eggertsen, *Anal. Chem.* 30, 1387–1390 (1958).
61. L. S. Ettre, *Paper, 11th Pittsburgh Conference on Analytical Chemistry and Applied Spectroscopy, March 1, 1960*.
62. *Instructions, Perkin-Elmer-Shell Model 212 Sorptometer*, Perkin-Elmer Corp., Norwalk, Conn., 1961.
63. H. W. Daeschner and F. H. Stross, *Anal. Chem.* 34, 1150–1155 (1962).
64. J. H. Atkins, *Anal. Chem.* 36, 579–583 (1964).
65. T. C. McFearin, *Rubber Age (N.Y.)* 91, 611–613 (1962).
66. J. C. Krecji and C. H. Roland, *Paper, Fall Meeting, American Chemical Society, Division of Rubber Chemistry, Cleveland, Ohio, 1965.*
67. W. B. Innes, *Anal. Chem.* 23, 759–763 (1951).
68. *Instruction Manual*, C. W. Brabender Instruments, Inc., South Hackensack, N.J.
69. Sljaka, et al., Cabot release for D-24 ASTM Meeting, Committee on Carbon Black, May 1, 1963, American Society for Testing and Materials, Philadelphia, Pa.
70. E. R. Eaton and J. S. Middleton, *Rubber World* 152, 94–100 (1965).
71. C. W. Sweitzer and W. C. Goodrich, *Rubber Age (NY)* 55, 469 (1944).
72. A. Voet and W. M. Whitten, Jr., *Rubber World*, 148 (5), 32–36 (Aug. 1963).
73. A. I. Medalia and R. L. Sawyer, *Proc. 5th Carbon Conference, New York, 1961*, Pergamon Press Inc., New York, 1963, pp. 563–584.
74. A. I. Medalia, *Rubber Age (N.Y.)* 93 (4), 580–586 (July 1963).
75. *ASTM D 1680-60 (1968), Test for Volatile Content of Carbon Black*, American Society for Testing and Materials, Philadelphia, Pa., 1968
76. *ASTM D 1512-60 (1968), Test for pH Value of Carbon Black*, American Society for Testing and Materials, Philadelphia, Pa., 1968.
77. *ASTM D 1513-60, Method of Test for Pour Density of Pelleted Carbon Black*, American Society for Testing and Materials, Philadelphia, Pa., 1966.
78. R. P. Rossman and W. R. Smith, *Ind. Eng. Chem.* 35, 972–976 (1943).
79. *ASTM D 1506-59 (1968), Test for Ash Content of Carbon Black*, American Society for Testing and Materials, Philadelphia, Pa., 1968.
80. *ASTM D 1509-59, Method of Test for Heating Loss of Carbon Black*, American Society for Testing and Materials, Philadelphia, Pa., 1966.
81. *ASTM D 1619-60, Method of Tests for Sulfur Content of Carbon Black*, American Society for Testing and Materials, Philadelphia, Pa., 1966.
82. T. Battye, R. Johnson, and H. C. Wilkinson, *J. Appl. Chem.* 4, 119–123 (1954).
83. *ASTM D 1514-60, Method of Test for Sieve Residue from Carbon Black*, American Society for Testing and Materials, Philadelphia, Pa., 1966.
84. B. B. Bauminger and F. C. J. Poulton, *Analyst* 74, 351–360 (1949).
85. T. A. Purifoy, "Analyses for Percent Carbon Black in Rubber by Reduced Pressure Pyrolysis," *Paper, Meeting American Chemical Society, Division of Rubber Chemistry, Miami, Fla., May 4–7, 1965.*
86. I. M. Kolthoff and R. G. Gutmacher, *Anal. Chem.* 22, 1002 (1950).

B. Schubert
Columbian Carbon Company
F. P. Ford
Columbian Carbon Company
F. Lyon
Columbian Carbon International

(12)   Related Proceedings Appendix

None.